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Design of a proportional counter and its application to Tb156 radiation

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DESIGN OF A PROPORTIONAL COUNTER AND ITS APPLICATION TO Tb^{156} RADIATION

by

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Dean of Graduate College

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I. INTRODUCTION AND SURVEY OF PREVIOUS WORK

Irradiation of matter in the external bremsstrahlung beam from an electron synchrotron ordinarily yields a level of radioactivity which is low compared to that readily induced by heavy charged particle bombardment in a cyclotron or exposure to the high neutron flux in a nuclear reactor. Of the devices available for the investigation of the energy spectra of electrons and quanta from such synchrotron-produced sources, magnetic beta-ray spectrometers and crystal diffraction instruments are capable of excellent resolution but are handicapped by their requirement for relatively intense sources; while crystal scintillation spectrometers lack the desired energy resolution, particularly at energies below a few hundred kev.

Gas-filled proportional counters offer certain advantages in this respect. Although used for many years as radiation detectors (1,2,3,4, 5), the full extent of their usefulness, particularly in the low energy region, was first indicated by Kirkwood et al. (6) and Curran et al. (7) less than ten years ago. Since then, considerable effort has been expended (8,9,10,11) along experimental lines dictated by the following unique characteristics of proportional counter spectrometers:

a. Adaptability to the use of gaseous sources for the study of soft electrons.

b. Essentially 4π geometry for sources placed on internal probes, thus allowing the use of weak sources.

c. Ability to accommodate sources spread over several square centimeters of a probe or over the entire cathode surface if necessary,

thus making possible the study of sources of low specific activity.

d. A quantum detection efficiency far less than that of scintillation counters, but much better resolution at energies below about 100 kev. The attainable precision of measurement of energies of x-rays, soft gamma-rays and conversion electrons is exceeded only by crystal diffraction methods and high-resolution magnetic beta-ray spectrometers.

e. Sufficient portability to be located near an accelerator for measurement of short-lived activities which must be transported rapidly from irradiation position to counting location.

To exploit these useful characteristics, a large cylindrical proportional counter has been designed and constructed. The counter is intended to operate at gas pressures up to ten atmospheres and has an active counting volume of just over one and one half cubic feet. It is equipped with beryllium and aluminum side windows for the admission of electromagnetic radiation from external sources and a source probe operating through a vacuum lock which enables internal sources to be inserted or withdrawn without disturbing the counting gas. A strong axial magnetic field is available to constrain energetic electrons from internal sources to the active counting volume, and "field tubes" as proposed by Cockroft and Curran (12) have been employed to reduce end effects.

After thorough testing, this counter has been applied to the investigation of the radiation from Tb^{156}, which was produced in the synchrotron bremsstrahlung beam by the reaction $Tb^{159}(\gamma,3n)Tb^{156}$.

Considerable disagreement exists in the literature regarding the radiation from Tb^{156}. Krisberg et al. (13) detected a 5.5-day activity following the bombardment of gadolinium with either neutrons or deuterons but were unable to make an unambiguous mass assignment. The deuteron-produced activity was found by Butement (14) to consist of two components: a 6.75-day activity in Tb^{161} and a 5.9-day activity in some other terbium isotope, probably Tb^{156}, Tb^{157}, or Tb^{158}. In 1948, Wilkinson and Hicks (15,16,17,18) undertook a systematic study of neutron-deficient rare-earth isotopes produced by proton, deuteron, and alpha-particle bombardment of the various rare-earth elements. They employed end-window G-M counters, magnetic deflection to distinguish between positive and negative electrons, energy determination by absorption in beryllium, aluminum and lead, and the then newly developed technique of ion-exchange resin column separation of the rare-earth elements. After bombardment of europium with 10-Mev alpha-particles, a 5.0 ± 1-hour activity was observed (18) and assigned to Tb^{156} on the basis of yield. These investigators reported the decay of Tb^{156} to take place mainly by orbital electron capture with less than 25 per cent positron branching. The positrons were found to have a maximum energy of approximately 1.4 Mev. No negative electrons were observed in a beta-ray spectrometer, but aluminum absorption showed hard electrons of about 1.3 Mev, soft electromagnetic radiation (presumably x-rays) of about 6.3 kev, and a hard quantum radiation background. Insufficient activity was available for a lead absorption measurement of gamma-rays present.

Handley and Lyon (19) have surveyed the activities in neutron-

deficient isotopes of terbium produced by proton bombardment of enriched isotopes of gadolinium. In Tb^{156}, they observed activities with half-lives of 5.2 days and 5.5 hours, along with "a small fraction of longer-lived component". A ratio of 80 to 1 (5.5-hour to 5.2-day) was observed for these activities, which ratio was independent of enrichment and bombarding particle energy, thus leading to the conclusion that the two activities were isomeric. They were assigned to Tb^{156} because they were produced by 22.4-Mev protons on Gd^{158} and by 9.5-Mev protons on Gd^{157} containing 7.31 per cent Gd^{156}. By aluminum absorption measurements, the 5.5-hour activity was identified as a single beta group with an end point energy of .14 Mev. After the 5.5-hour activity had died out, other aluminum absorption data led to identification of the 5.2-day activity as consisting of two beta groups with maximum energies of .6 and .2 Mev, K x-rays from gadolinium and ten gamma-rays. It is of interest to note that beta decay of Tb^{156} would lead to Dy^{156} and K-capture would lead to Gd^{156}. The K/β^- ratio was concluded to be large, and no annihilation radiation was detected, indicating little or no β^+ branching in competition with the K-capture. With a proportional counter, L x-rays from dysprosium with a 5.5-hour half-life were observed, indicating a highly converted weak gamma-ray. As the L x-ray peak decayed with this 5.5-hour half-life, the energy of the peak shifted from that of dysprosium to that of the L x-ray of gadolinium, which peak then continued to decay with a half-life of 5.2 days. Weak conversion electrons in the short-lived activity were detected with a windowless proportional counter.

Mihelich and Harmatz (20) have reported a 5.5-hour activity in Tb^{156} with an energy of 88.2 kev. This activity was produced by proton bombardment of enriched gadolinium followed by ion-exchange separation and was attributed to an isomeric transition in Tb^{156}. The assignment was based on variations in target enrichment and the observation that the activity was genetically related to transitions known to take place in Gd^{156}.

Mihelich et al. (21) have undertaken a survey of the radioactive decay chains in the rare-earth region. They have bombarded enriched gadolinium with protons ranging in energy from 12 Mev to 22 Mev, using various enrichments and separating the reaction products by ion exchange. Resultant activities were examined in a standard conversion electron spectrograph in which successive exposures were made to allow photometric determination of the decay rates of individual lines. They drew the following conclusions regarding Tb^{156}:

a. There is an E3 isomeric transition of 88.4 kev with a 5.5-hour half-life.

b. The ground state activity is observed to grow and then decay.

c. No conclusive evidence was found for a transition de-exciting the first excited level in Dy^{156}, which might be fed by β^- decay of Tb^{156}.

d. Scintillation counter data are in good agreement with Handley and Lyons (19), except that not all of the transitions reported by them with energies between 1 Mev and 2 Mev are believed to occur in isotopes of mass 156.

e. It is inferred that Tb^{156} decays by K-capture to Gd^{156}. Subse-

quent transitions in Gd^{156} were shown to exhibit a 5.6-day half-life and energies of 89.10, 111.9, 155.2, 199.4, 262.7, 296.7, 356.6, and 422.2 kev.

In confirmation of the assignment of two of these levels to Gd^{156}, Mihelich pointed out that Heydenburg and Temmer (22) have reported Coulomb excitation of an 89-kev level in Gd^{156}; and Church and Goldhaber (23) have observed capture gamma-rays with energies of 88.8 kev and 198.7 kev in the same isotope.

Most recently, Dillman et al. (24,25) have reported gamma-rays with energies of 88, 130, 200, 250, 340, 540, 620, 1210 and 1410 kev, plus weak radiation of other energies from Tb^{156} produced by irradiation of terbium oxide with bremsstrahlung of 240 Mev maximum energy. Assignment of this activity to Tb^{156} was based on a comparison of these results and observed half-lives of 5.5 hours and 5.2 days with the published data of Handley and Lyon (19). At least three beta groups leading to excited states of the daughter Dy^{156} nucleus were detected. By gamma-gamma and beta-gamma coincidence measurements, the 200, 540, 1210, and 1410 kev gamma-rays were grouped together and observed to follow beta decay of Tb^{156}; while other gamma-gamma and gamma-x-ray coincidence studies associated the 130, 250, 340, and 620 kev radiations with each other and led to their identification as following orbital electron capture. The earlier assignment (20) of the 88-kev transition to an isomeric state in Tb^{156} was confirmed by the absence of coincidences involving this radiation.

In an attempt to resolve some of these apparent discrepancies,

several samples of highly purified terbium metal and terbium oxide (Tb_4O_7) have been irradiated in the external 64-Mev bremsstrahlung beam of the Iowa State College electron synchrotron in order to produce Tb^{156} from the single stable isotope of terbium by the reaction $Tb^{159}(\gamma,3n)Tb^{156}$. The irradiated samples have been examined in several ways. Half-life measurements have been made on the total activity with an end-window G-M counter and on selected spectral regions with a scintillation spectrometer and a multi-channel analyzer (26). Electrons have been sought with a large proportional counter and an anthracene scintillation spectrometer. The spectrum of electromagnetic radiation has been examined with a scintillation spectrometer employing a NaI(Tl) crystal; and finally, a study was made of gamma-rays detected by the scintillation counter in coincidence with x-rays detected by the proportional counter.

II. INSTRUMENTATION

A. Large Proportional Counter Spectrometer

1. Theoretical considerations

Proportional operation of a gas-filled counter of fixed configuration is characterized by a ratio, E/p, of electric field strength to gas pressure which is sufficiently great at some region in the counter that an electron liberated within the sensitive volume gains enough energy during one free path of its motion under the influence of the applied field to produce another ion pair by collision with a molecule of the counting gas. With a smaller value of E/p, mere separation and collection of the components of the ion pairs takes place and the counter operates as an ionization chamber; while an increase of E/p beyond the upper limit of proportionality leads to operation in the Geiger region, in which the amplitude of the counter's output pulse is independent of the number of initial ion pairs produced by the original ionizing event.

The phenomena involved in the multiplication process have been described in detail in the literature (2,3,4,5,9,10), but several features of the theory deserve particular attention in view of the effort here to achieve high resolution, the contemplated use of this counter in coincidence experiments and the desire to make available data which may aid in the future design and operation of similar instruments.

An important consideration in coincidence application of a gas-filled counter is the delay which occurs between the passage of the incident radiation through the sensitive volume and the beginning of the out-

put pulse. Since this delay depends upon the electron drift velocity as a function of E/p for a given gas mixture and upon the initial distance of each primary ion pair from the anode, its exact calculation is not practical; but, an estimate is of value in indicating the order of magnitude of the delay to be expected.

The electric field strength at a radius r inside a cylindrical counter with a coaxial anode is

$$E = \frac{V_o}{r \ln(b/a)} , \qquad (1)$$

where V_o is the voltage at which the anode is maintained with respect to the surrounding cylindrical cathode and a and b represent the radii of the anode and cathode, respectively. With the help of Equation 1 and published measurements (27,28) of electron drift velocity as a function of E/p, the drift velocity v can be expressed in terms of r for a given counter with fixed values of a, b, p, and V_o. The time required for an electron to drift in to the anode is then given by

$$t_d = \int \frac{ds}{v} , \qquad (2)$$

in which ds is an element of the radial path along which the electron may be imagined to drift if deflections by collisions are ignored and where the integration is carried out over the drift distances between the electron's point of origin and the surface of the center wire. In Figure 1, using the data of English and Hanna (27), 1/v has been plotted against s (measured from the cathode) for the case of a counter with a = .0025 inches (6.35×10^{-3} cm), b = 6 inches (15.24 cm), V_o = 3000 volts and

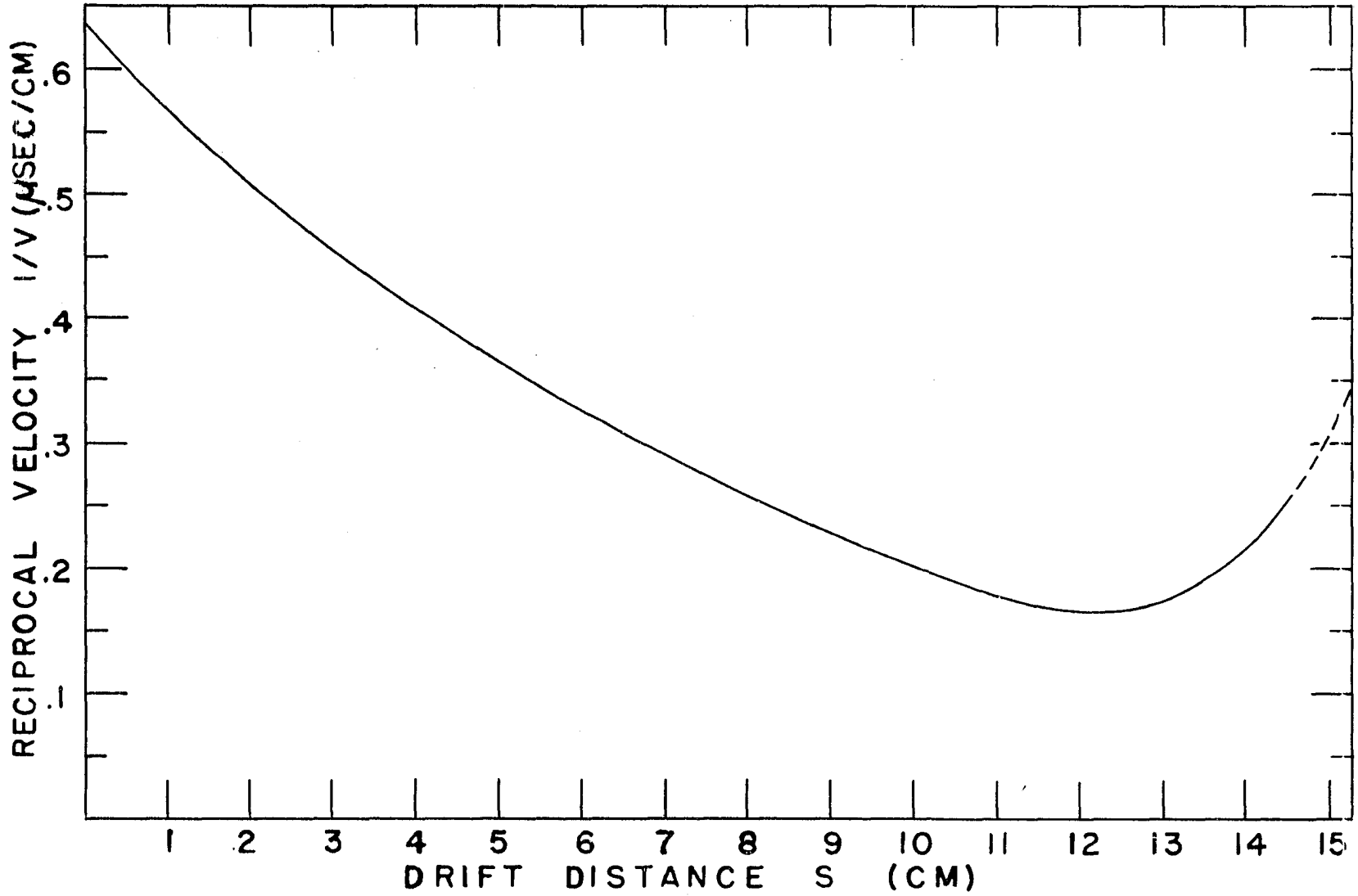

Figure 1. Curve for determination of electron drift time

p = 720 psi of a mixture of 90 per cent argon and 10 per cent methane. By measuring the total area under this curve, the extreme delay expected for an electron originating at the cathode is seen to be approximately 5 microseconds. The lower limit of the drift time, of course, is substantially zero for an electron formed very near the center wire; but the probability of such an event is small. Thus an average initial delay of the order of a few microseconds is to be anticipated.

At the end of the electron's drift path, in the high-field region very near the anode, the electron avalanche occurs which gives rise to the characteristic gas multiplication factor A, frequently referred to as the gas amplification or gas gain. Rose and Korff (3) have derived an expression for this factor,

$$A = \exp \left\{ 2(fNCaV_o)^{1/2} \left[(V_o/V_t)^{1/2} - 1 \right] \right\} , \quad (3)$$

in which f is a quantity characteristic of the filling gas, N is the number of molecules per unit volume of gas, C is the capacitance per unit length of the counter, a is the anode radius, V_o is the counter voltage and V_t is a proportional threshold voltage obtained by extrapolating the linear region of the curve of ln A versus V_o to A = 1. Unfortunately, although Equation 3 holds well over a reasonably large range of A, its utility in design calculations is greatly diminished by the fact that V_t must be determined experimentally by measurements of A for the counter in question. Rose and Korff have reported good agreement between theory and experiment for polyatomic gases such as methane at gas gains up to 10^4, but Korff (4) points out that this relationship breaks down for

values of A greater than approximately 10^2 in monatomic and diatomic gases, presumably due to the creation of ultraviolet photons during the avalanche and production of electrons at the cathode by positive ion bombardment.

As pointed out by Rossi and Staub (5), A is unchanged if, for constant p and a, V_o and b are changed so as to maintain a constant field $E = V_o/r \ln(b/a)$ near the center wire. Also, if a and b are multiplied by a constant k, p is divided by k and V_o is kept unaltered, A is again constant; since the energy acquired by an electron in one mean free path and the number of mean free paths within a distance of the order of a, where multiplication occurs, are both unchanged. Accordingly, the gas gain may be expressed as

$$A = A\left(\frac{V_o}{\ln(b/a)}, pa\right). \qquad (4)$$

Whenever measurements of A as a function of V_o are available for a counter of known configuration at various pressures, this relationship allows an estimate to be made of A in other counters employing the same gas mixture but with different values of a and b. Unfortunately, such extensive measurements were not available when the counter under consideration herein was being designed, but gas gain data obtained during the evaluation of this counter, which are presented in Table 2 later in this chapter, may be of value in future designs of similar devices.

The statistical fluctuations which occur in the formation of the initial ion pairs and in the subsequent electron avalanches during the multiplication process are of interest because of their limiting influ-

ence on the inherent resolution of the proportional counter. The statistical broadening of the line which ideally might be expected in an energy spectrum when monoenergetic radiation impinges on the counter may be described in terms of the variance, $v_t = \overline{N^2} - \bar{N}^2$, in the total number of electrons N produced by both initial ionization and subsequent cascade multiplication following the passage of one original quantum or fast particle through the counter gas.

Since all events are independent at each stage in the multiplication process, v_t may be expressed as a sum of two components, one due to fluctuations in the number of original ion pairs produced and the other due to fluctuations in the gas gain. Such an expression has been derived by Frisch (29):

$$v_t = v_o \, m_A^{\,2} + v_A \, m_o \, . \qquad (5)$$

In this equation, m_o is the mean value of the number of ion pairs initially liberated by the incident radiation, m_A is the mean number of electrons produced in the avalanche by a single ionization electron, and v_o and v_A are the variances in these quantities, respectively. A similar result has been obtained by Palmer and Laslett (30) in the treatment of the multiplicative processes in the crystal and photomultiplier of a scintillation counter.

Since m_A is simply the mean value of the gas gain or gas multiplication factor A; and since

$$m_o = \frac{E_i}{W} \, , \qquad (6)$$

where E_i is the energy of the incident particle or quantum and W is the energy required to produce one initial ion pair, all of which are subject to more or less direct measurement; a determination v_t awaits only the evaluation of v_o and v_A.

With the assumption that the probability of ionization is a function only of the distance of the electron from the center wire, Frisch also has obtained theoretically the expression

$$v_A = m_A^2 - m_A \ , \tag{7}$$

from which it may be seen that the relative or fractional variance in the number of electrons resulting from one initial electron,

$$\frac{v_A}{m_A^2} = 1 - \frac{1}{m_A} = 1 - \frac{1}{A} \ , \tag{8}$$

assumes a distressingly large value of approximately unity whenever the gas gain is appreciable. However, if Equation 7 is substituted into Equation 5,

$$v_t = v_o\, m_A^2 + m_A^2\, m_o - m_A\, m_o \ , \tag{9}$$

and the relative variance in the total number of electrons resulting from an avalanche started by m_o electrons becomes

$$\frac{v_t}{(m_A\, m_o)^2} = \frac{v_o}{m_o^2} + \frac{1}{m_o} - \frac{1}{m_A m_o} = \frac{v_o}{m_o^2} + \frac{1}{m_o} - \frac{1}{A m_o} \ . \tag{10}$$

In Equation 10, the third term on the right may be neglected, since

ordinarily $\bar{A} \gg 1$ for proportional counter operation. The first term on the right gives the relative variance in the original number of ion pairs; and the second term, representing the contribution of the multiplication process to the total relative variance, is seen to be inversely proportional to the mean value of the number of original ion pairs. Since m_o is usually quite large (a 30-kev electron which expends all its energy in an argon-filled counter will produce more than 10^3 original ion pairs), the broadening effect of the cascade process on the final distribution is not at all disastrous.

In fact, if a normal Poisson distribution is assumed in the initial ionization, $v_o = m_o$, and Equation 10 becomes

$$\frac{v_t}{(m_A\, m_o)^2} = \frac{2}{m_o} \quad ; \tag{11}$$

so the effect of the avalanche is merely to double the relative variance in the number of initial ions.

The assumption of a Poisson distribution may be justified when the incident radiation loses only a small fraction of its energy in the counter, as in the case of transit by a cosmic ray particle; but in the present application, the radiation is expected to remain in the counter until it loses all of its energy, so that if it does not ionize as expected in a given length of path, it simply retains energy with which it may ionize later. Thus one might suspect that $v_o < m_o$. Confirming this suspicion, West (10) displays experimental data in agreement with Fano's calculation (31) that v_o should lie in the range $m_o/2 > v_o > m_o/3$.

A slightly different value has been obtained by Curran et al. (11, 32), who have measured the fluctuations in the multiplication process alone by releasing single low-energy photoelectrons in a counter and have concluded that the two components of the relative variance are roughly equal and that each has a magnitude of about $.67/m_o$.

Finally, the anticipated shape of the output voltage pulse from the counter must be examined. After completion of the avalanche, which can only occur very close to the center wire (within a distance of the order of one anode radius), the electrons move very quickly to the wire because of their high mobility; but the consequent change in potential of the wire is negligibly small, since the electrons do not have far to travel and the slowly-moving positive ions are still in the immediate vicinity. The major portion of the proportional counter pulse results from the outward motion of these ions through the high-field region toward the cathode. If the counter is considered simply as a capacitor, it is clear that the final pulse height is given by

$$P_f = \frac{A\, n_o\, e}{C} \quad , \tag{12}$$

where A is the gas gain, n_o is the number of initial ion pairs formed by the incident radiation, e is the electronic charge and C is the capacitance of the counter and the associated input circuit to its amplifier. Wilkinson (9) and others (33) have shown by energy considerations that the pulse height due to the positive ions as a function of time is

$$P(t) = \frac{A\, n_o\, e}{2C\, \ln(b/a)} \ln\left[1 + \frac{t}{a^2 \ln(b/a) \;/\; (2V_o K/p)}\right] . \tag{13}$$

For the same representative values of the counter parameters used in plotting Figure 1, and setting $K/p = 3$ cm^2/volt sec, in accordance with the values given by Wilkinson (9) for the mobility of positive ions in argon, Equations 12 and 13 may be used to show that although the time required for the pulse to reach its maximum value (collection of the positive ions at the cathode) is measured in thousands of microseconds, it reaches half of maximum in about 40 microseconds and one-fourth of maximum in less than one microsecond. Consequently, the pulse may be clipped to a useable length of a few microseconds by subsequent pulse-shaping circuits without sacrificing proportionality and without reducing the pulse height inordinately. Of course, in the case of relatively energetic incident radiation, such as electrons with energies of several hundred kev and correspondingly long ionizing paths, care must be taken to keep the clipping time long enough so that electrons formed far from the center wire have an opportunity to contribute to the output pulse.

2. Design and construction

Four main considerations were held foremost in the design of the proportional counter:

a. Ability to accept electrons with energies up to approximately one Mev.

b. Rugged construction to insure reliability of operation.

c. Sufficient versatility to allow its use with a wide variety of sources, both external and internal.

d. Highest possible resolution.

The proportional counter, a sectional view of which is shown in

Figure 2, consists basically of a stainless steel cylinder 12 inches in inside diameter and 36 inches long, with side ports through which radiation from external sources may be admitted, provision for the insertion of an internal source and a .005-inch diameter stainless steel anode wire down the center. Stainless steel was chosen for the construction of the cylinder because of its strength, its non-magnetic properties, its chemical inertness and the ease with which its machined inner surface can be kept clean and smooth. The stainless steel center wire appears, under a low-power microscope, to have a very smooth surface and uniform cross-sectional area. Its great strength makes its installation tremendously easier than that of, for example, a platinum wire of comparable size. In fact, stainless steel anodes .001 inch in diameter have been strung through the 36-inch length of the counter without mechanical difficulty.

The energy-handling requirement is responsible for the unusually large size of the counter. Even so, when the counter is filled with argon to its design limit of 10 atmospheres, an electron with an energy of only about 800 kev can penetrate the 4.5 inches of gas between the internal source probe and the nearest wall of the counter. Consequently, in order to reduce highly undesirable wall effects when working at high energies or low gas pressures, the further precaution has been taken of surrounding the counter with large water-cooled coils which, when carrying a current of 240 amperes, are capable of producing a fairly uniform axial magnetic field of approximately 2500 gauss throughout the sensitive volume of the counter. Figure 3 shows the counter in place inside the magnet coils. The axial field produced by these coils helps constrain to the

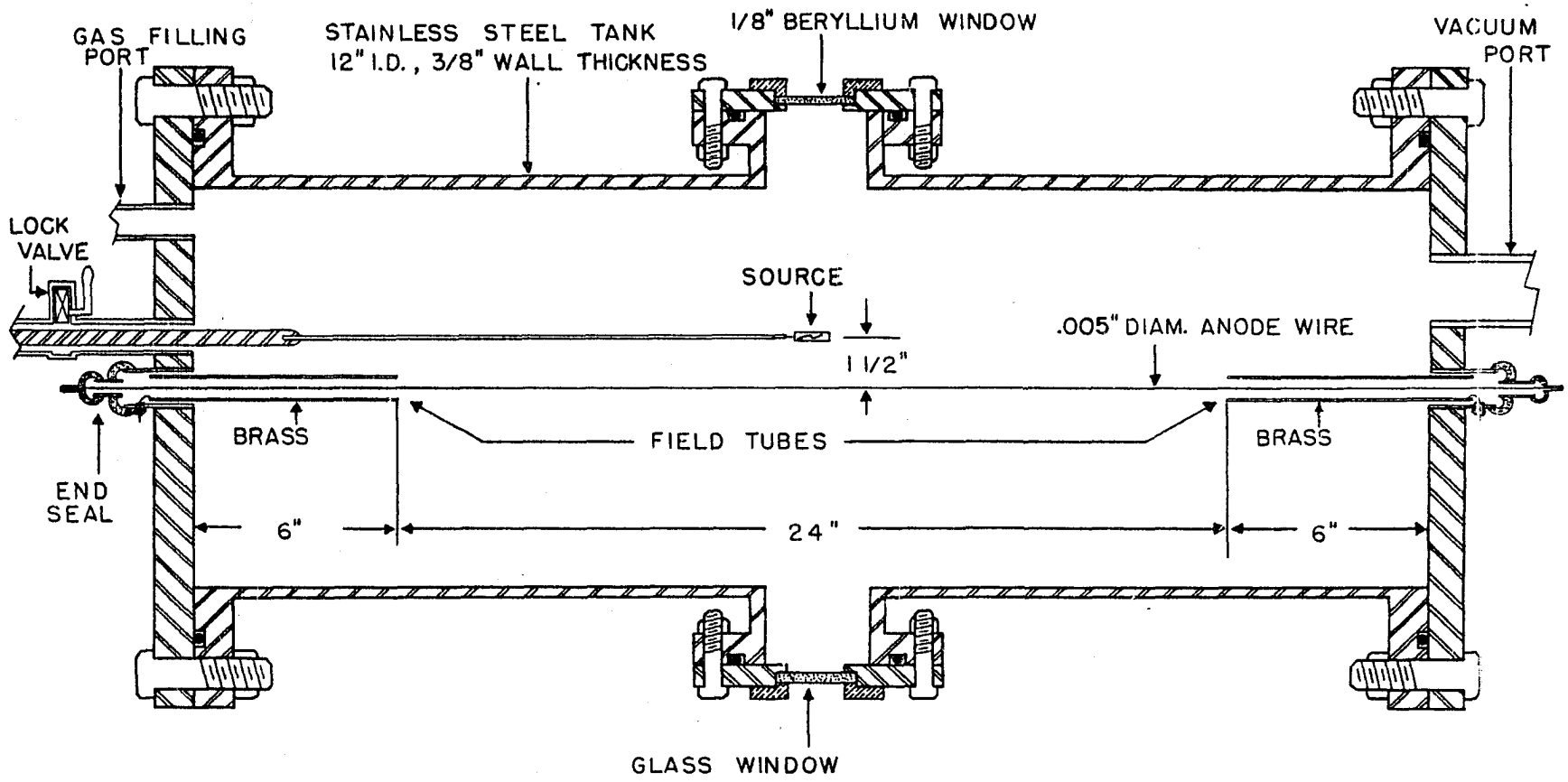

Figure 2. Longitudinal section of proportional counter

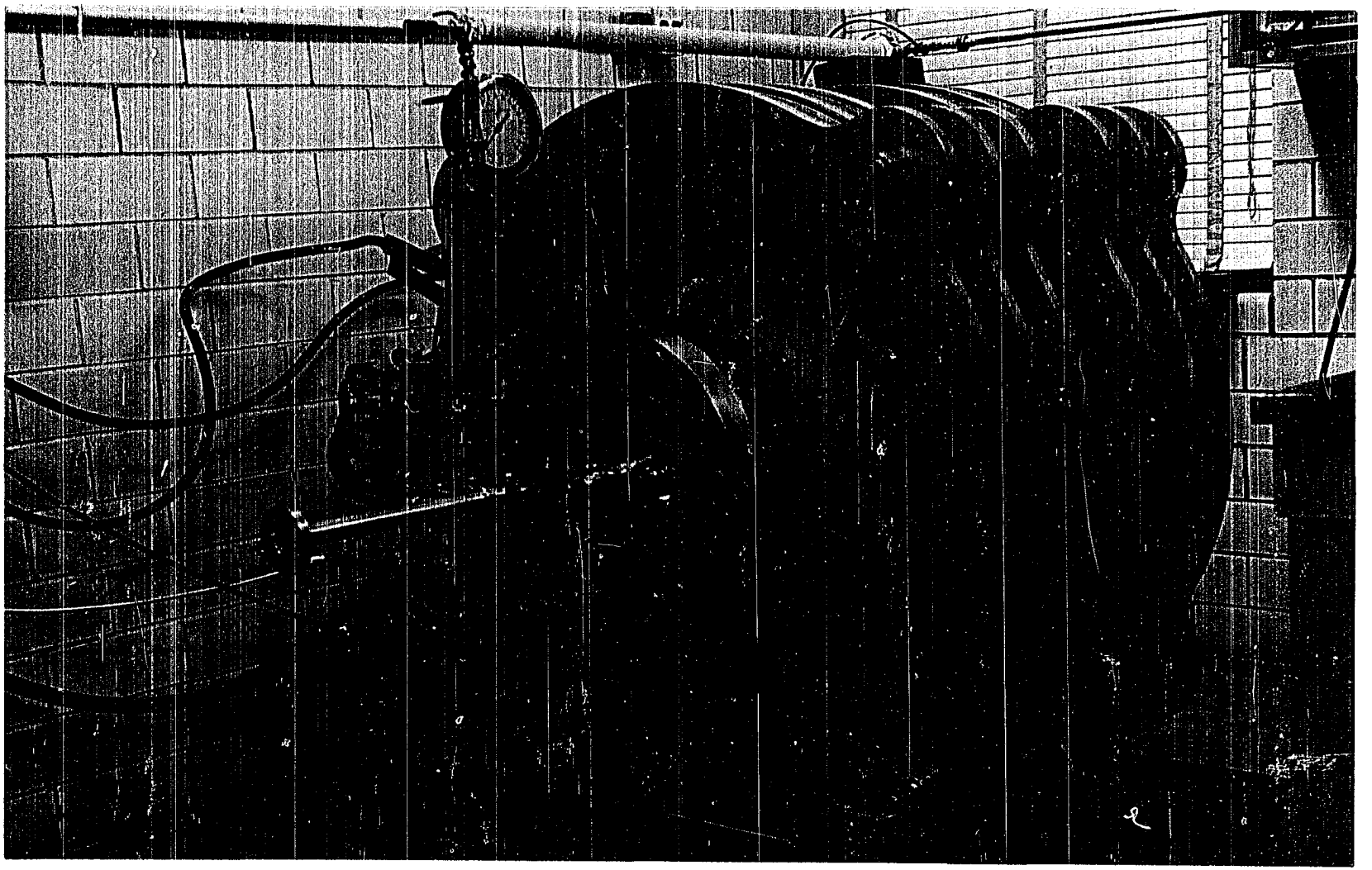

Figure 3. Proportional counter situated in magnet coils

sensitive volume those energetic electrons from an internal source which have an appreciable velocity component perpendicular to the axis of the counter, but those with a large velocity component parallel to the counter axis may still escape to the end walls. West (10) describes the magnetic field configuration which is necessary to confine an electron in three dimensions; but such a field, requiring two point-like pole faces, is not practical for a counter of this size. Consequently, small losses may be anticipated in dealing with electrons of high energy.

To avoid end effects due to regions of abnormally high or low electric field which occur at the end of the counter wire when the counter wire is either unshielded or surrounded by a guard tube at wire potential, "field tubes" of the type proposed by Cockroft and Curran (12) have been employed. When held at a potential equal to that existing in the body of the counter at the same radius, these tubes, which protrude for one counter radius (6 inches) into the counter, produce a field at the wire at this point which is the same as in the body of the counter.

Since soft electrons experience serious energy degradation in passing through even very thin windows, sources of such electrons should be located inside the sensitive volume of the counter. Also, for a source localized on a probe near the center of the cylinder, the counter presents a solid angle for detection of nearly 4π steradians, a further advantage when dealing with low-intensity sources. To allow the insertion of such an internal source, provisions have been made for a source probe which may be inserted into or withdrawn from the counter through a vacuum lock with an "O-ring" seal which prevents disturbance of the gas inside the

counter. The main features of this lock arrangement are illustrated in Figure 4. When inserted, the probe must be maintained at the proper potential corresponding to its distance from the axis of the cylinder in order to avoid distortion of the electric field near the anode.

As shown in Figure 1, two side ports are located midway along the counter. One of these ports usually contains a laminated glass window through which the interior of the counter may be viewed to check orientation and condition of internal sources, etc. The other contains a window of low -Z material (aluminum or beryllium) through which electromagnetic radiation from external sources may be admitted. Such an arrangement is most useful, in that it allows calibration of the counter with x-rays of known energies while an internal source is in place.

The end seals through which electrical connections are made to the anode, guard ring and field tubes are of unique design. These seals, one of which is shown in Figure 5, must be mechanically stable and strong enough to withstand 10 atmospheres of internal gas pressure, in addition to having sufficiently good insulating properties to endure the full impressed counter voltage of up to 5100 volts between anode (or the guard ring) and the grounded cylinder. They are fabricated from standard glass-to-metal seals in conjunction with a standard Cenco vacuum coupling as shown and have proven reliable, simple to construct and easy to install or remove.

Figure 6 is a schematic drawing of the vacuum system and gas filling apparatus connected to the proportional counter. Thorough evacuation is necessary to remove traces of oxygen and other residual gases which might cause deterioration of the counter's resolution through recombination or

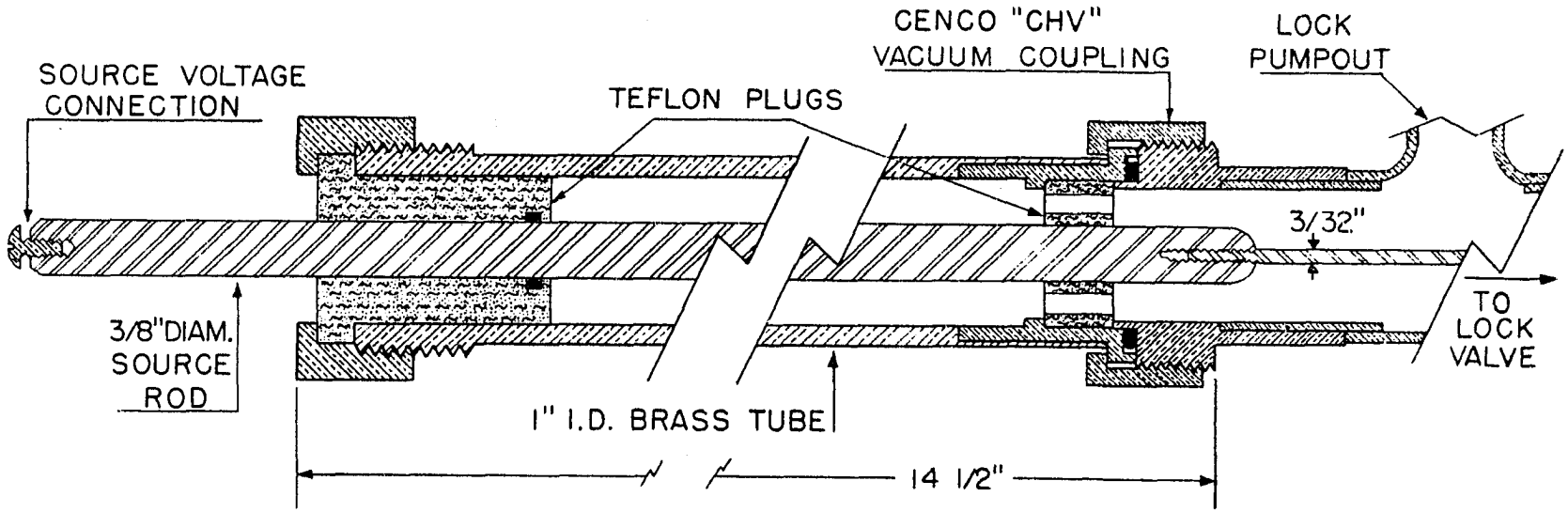

Figure 4. Longitudinal section of source lock with source rod in place

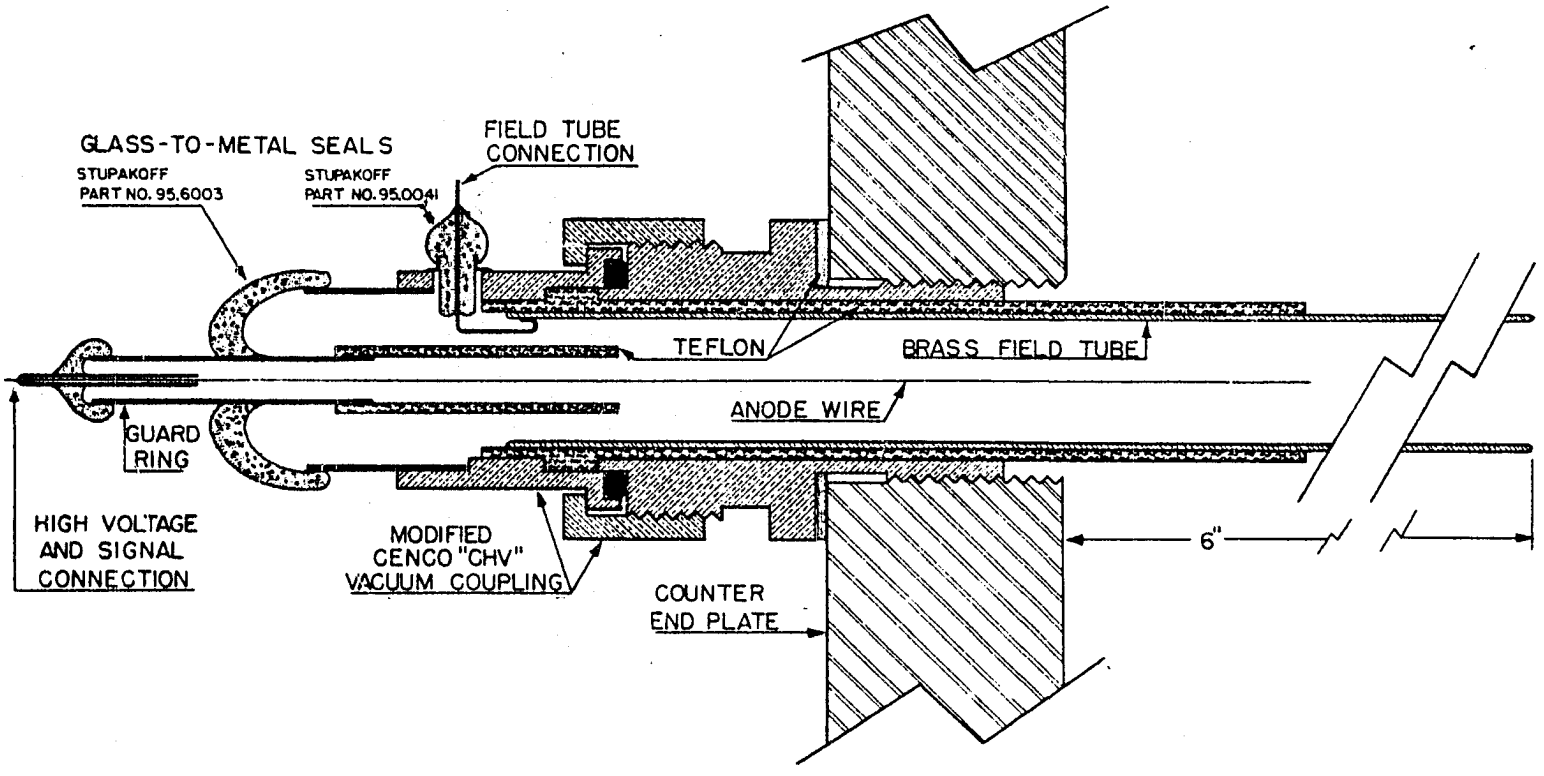

Figure 5. Longitudinal section of proportional counter end seal, showing electrical connections to anode, guard ring and field tube

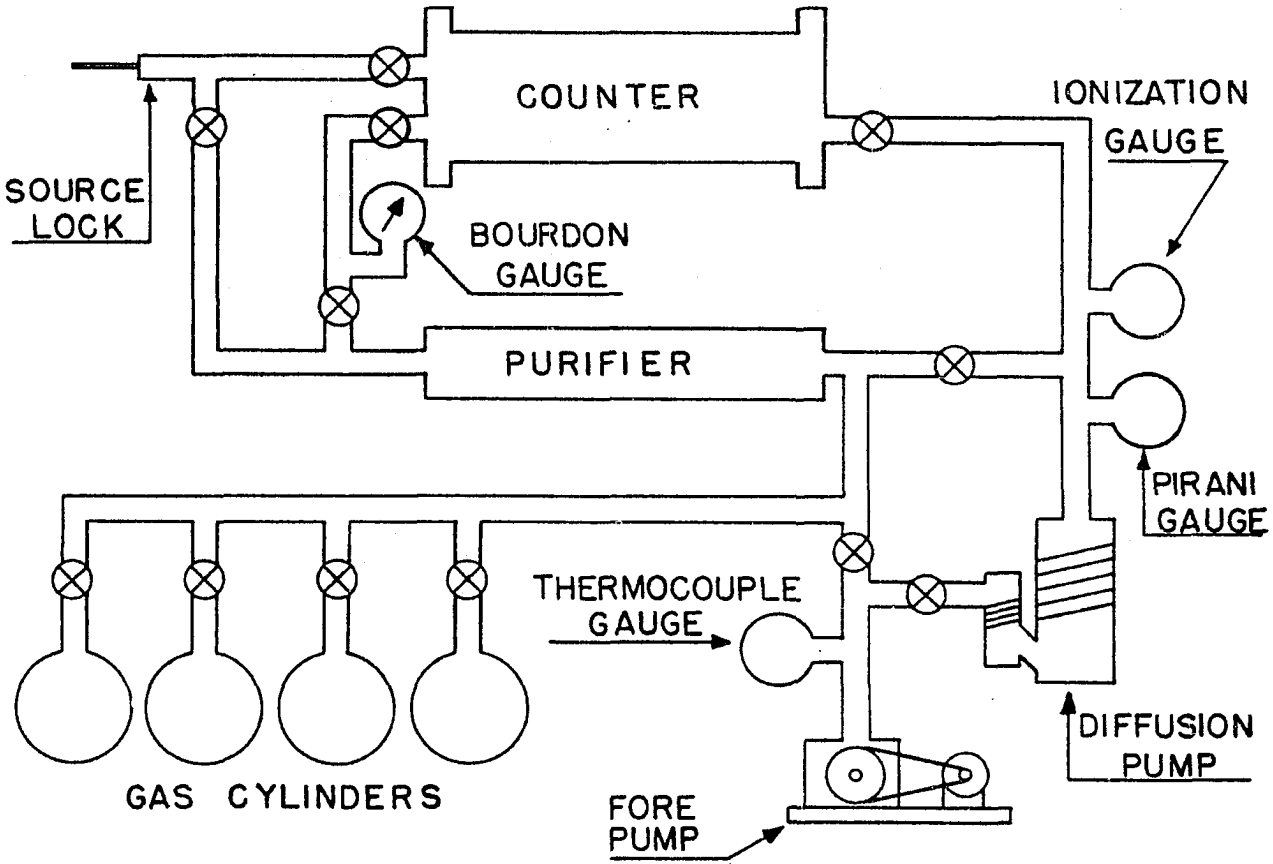

Figure 6. Block diagram of vacuum and gas filling systems for proportional counter

other unwanted effects. With an oil diffusion pump, the system can be evacuated to an ultimate vacuum of about three millimicrons of mercury without use of a cold trap. Provision has been made for the insertion of a trap if necessary, but to date the need for one has not been established. The vacuum gauges are quite conventional. A thermocouple gauge constantly monitors the fore vacuum, and a Pirani gauge and an ionization gauge are installed on the high-vacuum side of the diffusion pump. Examination of the valve arrangement indicated in Figure 6 will show how a source to be used internally can be inserted into the source lock, the lock evacuated and refilled to the proper pressure with counter gas and the source then thrust into the desired position inside the counter without loss or contamination of the gas already in the counter.

The gas manifold will accomodate four cylinders at one time. One cylinder contains helium for flushing the system, while the other three contain filling gases which may be mixed in any desired proportions.

Many gas-filled counters are equipped with gas purifiers to remove unwanted contaminants from the counting gas. Usually these involve passing the gas over trays of hot calcium chips, which are particularly effective in removing oxygen. However, the work of Gibbs, *et al.* (34) indicates that hot (500°C) copper should be equally suitable. Accordingly, a gas purifier was constructed by winding a spiral of copper around a long calrod-type heating element and supporting this along the center of a long asbestos-wrapped copper tube through which the filling gas passes on its way from the gas manifold to the counter. This purifier is clearly visible just above the magnet coils in the photograph of Figure 3.

3. Auxiliary electronic instrumentation

Since the gas amplification in a proportional counter of this type varies exponentially with the applied voltage, it is vital that the high voltage supply be exceedingly stable, well regulated and as noise-free as possible. Fortunately, such supplies are commercially available. One which has been used for most of the work with this counter is the Beva Model 301 power supply[1] with negative ground, output voltage variable from 1000 to 5100 volts and capable of delivering up to one milliampere. As specified, this power supply has been found to have regulation of .01 per cent for load and line variations, stability of .01 per cent for several hours and .1 per cent per day, and a noise and ripple content of less than .01 per cent.

Figure 7 shows how the high voltage is supplied to the center wire through a 20 megohm resistor and is also impressed across a string of fixed and variable resistors totaling 8.5 megohms. The two variable taps on the voltage divider thus formed supply the voltage required to maintain the field tubes and the source probe at the appropriate potentials. If the outer wall of the counter is grounded and a voltage V_o is applied to the center wire, these potentials are given by

$$V = V_o \frac{\ln(r/b)}{\ln(a/b)} , \qquad (14)$$

where a = radius of center wire, b = radius of counter, and r = radius at

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[1]Available from Beva Laboratory, 1640 Olden Avenue, Trenton, New Jersey.

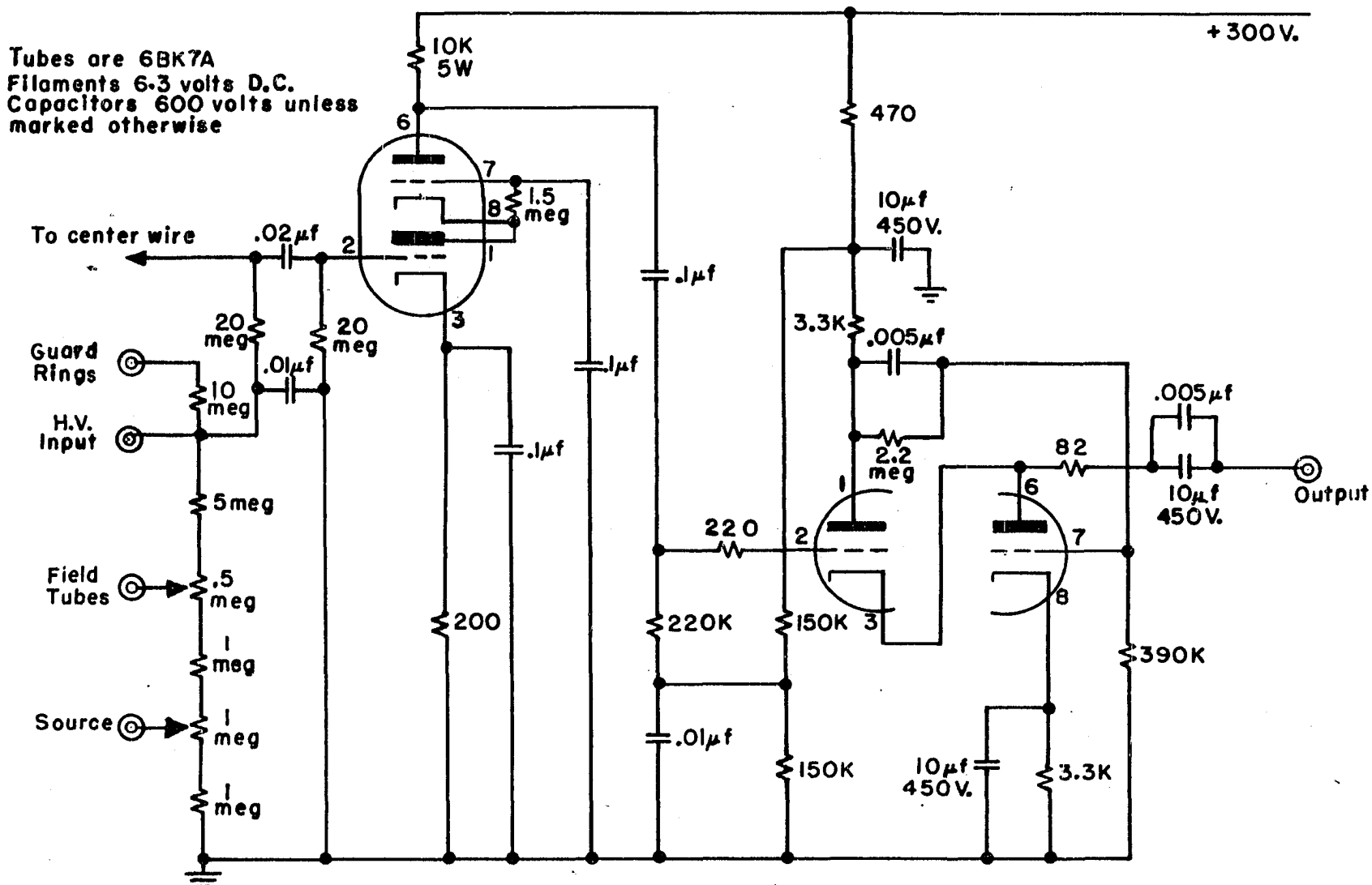

Figure 7. Circuit diagram of high voltage divider and preamplifier for proportional counter

which probe or field tube in located. Thus the potential is in each case directly proportional to the total counter voltage V_o, and once the variable taps on the high voltage divider shown in Figure 7 are correctly positioned, the source probe and field tubes are automatically maintained at the proper potential when V_o is varied. It will be noted that the guard rings, which draw no appreciable current, are held at the potential of the center wire. They serve simply as a protective device to preclude spurious pulses as a result of small leakage discharges which could occur between high voltage and ground across the lead-through insulators of the end seals.

Signal pulses are fed from the center wire to the input grid of the preamplifier through a .02-microfarad Glassmike capacitor which must be kept meticulously clean if spurious noise pulses due to leakage across its surface are to be avoided. From the pulse standpoint, the 10-$\mu\mu$f capacitance of the counter is shunted to ground by a 10-megohm resistor comprised of the two 20-megohm resistors in parallel, thus forming an input network to the preamplifier with a time constant of 100 microseconds to eliminate the extremely long tails of the proportional counter pulses. Further pulse shaping and much more drastic clipping takes place later in the linear amplifier.

The cascode input stage of the preamplifier shown in Figure 7 was selected because of its superiority in signal-to-noise ratio. (90 for the cascode arrangement as compared with 75 for a simple 6AK5 pentode.) A conventional cathode follower output stage employing the two sections of a 6BK7A tube connected in parallel was first tried but proved to have an

insufficiently low output impedance to drive the 50 feet of coaxial cable leading to the main linear amplifier, which was necessarily remotely located. The high voltage divider and preamplifier are built into a well-shielded subassembly which is attached to one end plate of the counter as shown in the photograph of Figure 8, thus maintaining the shortest possible lead length at low signal levels in order to minimize stray field pickup interference. The guard ring and field tube at the opposite end of the counter are supplied by shielded cables from this subassembly, as may also be seen in Figure 8.

The main amplifier has been selected with considerable care with respect to its stability, linearity and overload characteristics. Fortunately, as in the case of the power supply, suitable equipment is commercially available. The amplifier employed is a Cosmic Radiation Laboratories linear amplifier[2], Model 101, based on the design of Chase and Higinbotham (35), with a maximum gain of 3500, linearity of better than 1 per cent from 0 to 100 volts, boron carbon resistors with low temperature coefficients to reduce temperature instability, variable RC clipping, delay line pulse shaping and good overload properties. The non-overloading characteristic is attributable to the use of cathode-coupled amplifier stages (so-called "long-tailed pairs") in which the cathode is free to follow the grid swing on large positive pulses, thus preventing the flow of grid current and avoiding incorrect amplification of small pulses which closely follow pulses large enough to drive the amplifier out of its linear range. For most of the work reported here, the RC clipping

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[2]Available from Cosmic Radiation Laboratories, Inc., Patchogue, New York.

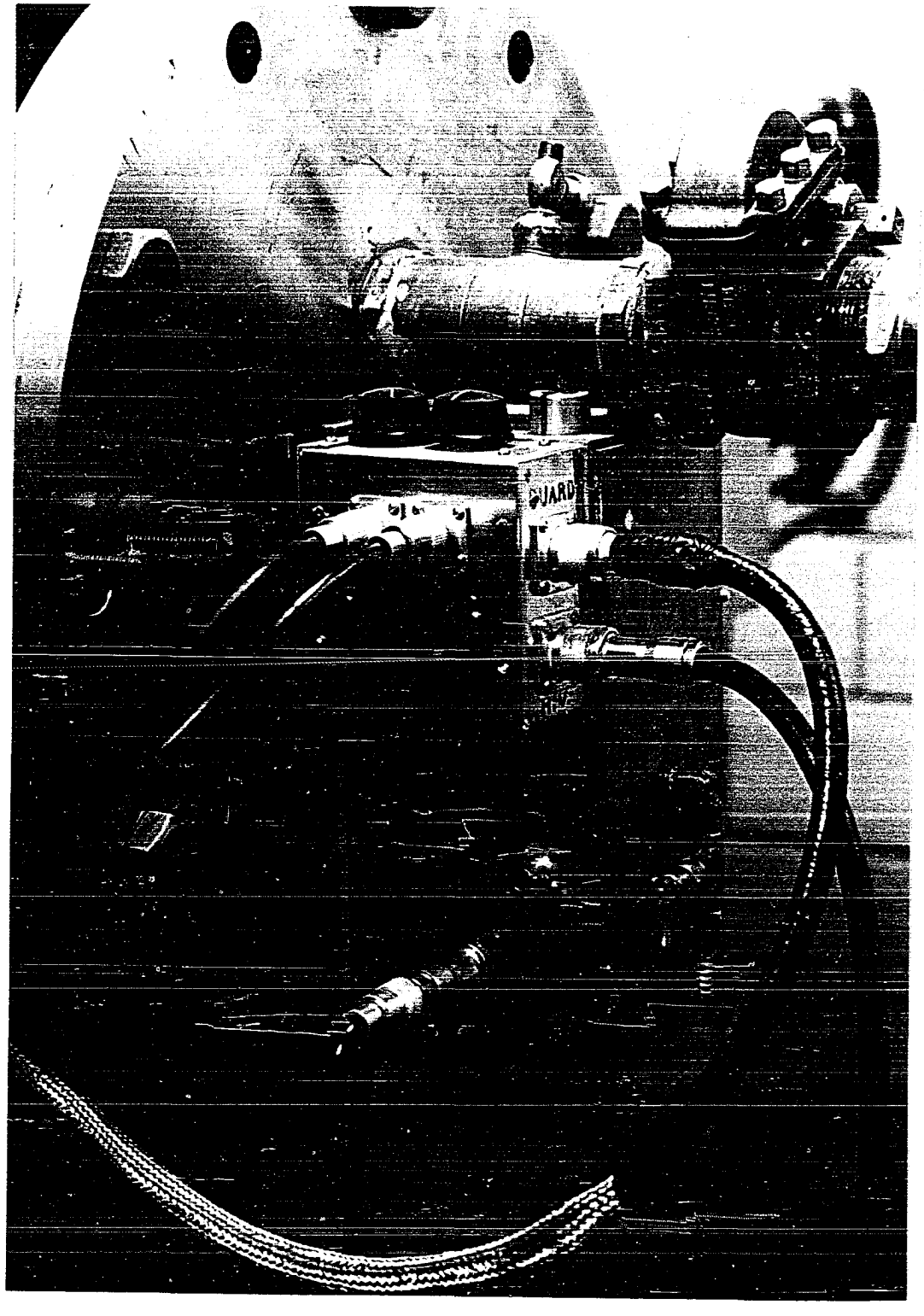

Figure 8. High voltage divider and preamplifier

was not used, but the length of the delay line in the amplifier was adjusted to give a relatively flat-topped output pulse about 10 microseconds in duration which may be subjected to pulse height analysis. The flat top is essential if the pulses are to be analyzed by the multichannel photographic analyzer mentioned later in this chapter.

A block diagram of the circuitry associated with the operation of the proportional counter is shown in Figure 9.

4. Test and evaluation

A number of tests have been conducted to evaluate the counter's capabilities. These have involved measurement of background level and its reduction by shielding, energy resolution of electrons and quanta, effect of the internal probe on resolution, effect of the magnetic field on resolution of high energy electrons, delays in the counter and the shape of the output pulse.

The background spectrum was examined with a single-channel analyzer and scaler, first with no shielding except that provided by the walls of the counter and subsequently with the surrounding coils in place and the space between them filled with a 4-inch thick layer of lead. The coils themselves provide 11.6 inches of copper shielding. The background spectrum was found to be reduced by approximately a factor of three throughout its extent by this shielding. No attempt has been made to incorporate relatively elaborate anti-coincidence arrangements of the type described by Curran (11) as being necessary in the examination of extremely low intensity radiation.

After considerable experimentation with different filling gases,

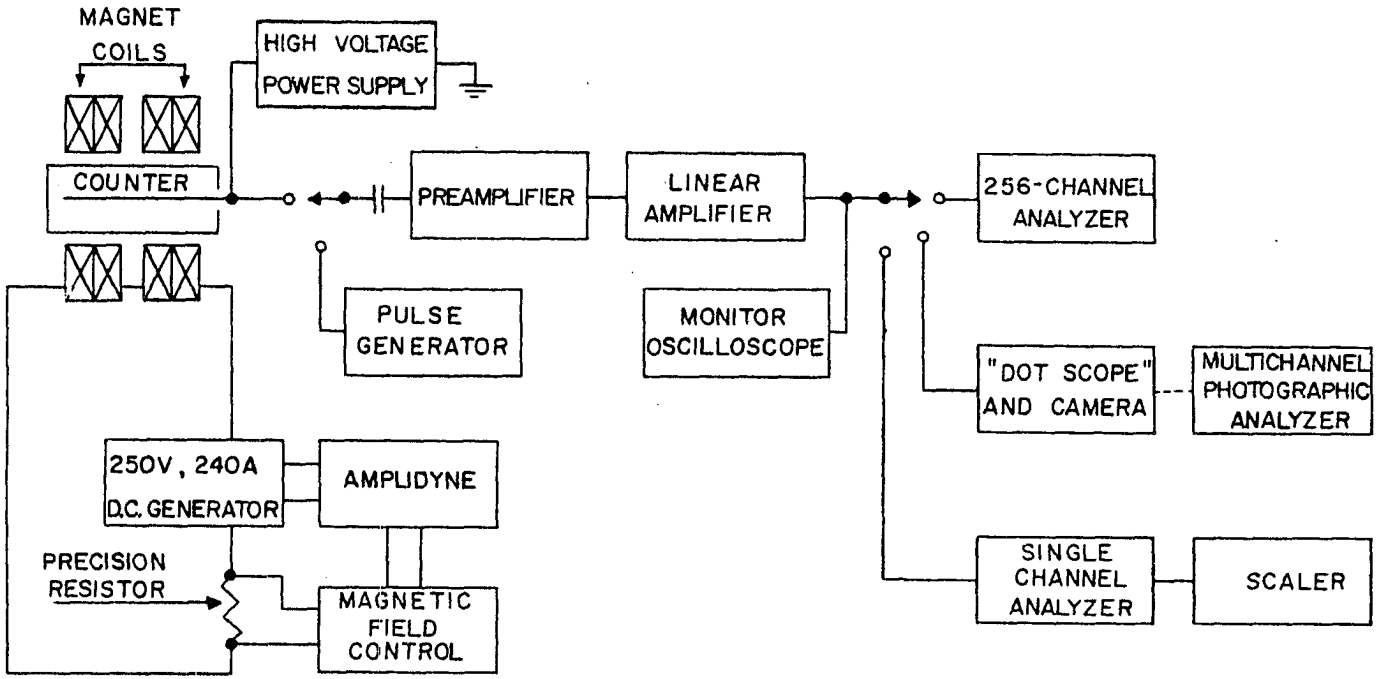

Figure 9. Block diagram of circuitry associated with the operation of the proportional counter

including argon, methane, argon-methane mixtures, argon-carbon dioxide mixtures and cyclopropane, a mixture[3] of 90 per cent argon and 10 per cent methane was adopted as most suitable for the majority of these experiments. The argon has a guaranteed purity of 99.9 per cent; and a representative analysis indicates a composition of 99.92 per cent argon, .08 per cent nitrogen and .001 per cent each of hydrogen and oxygen. The absolute maximum tolerance of hydrogen and oxygen is .002 per cent each. A representative sample of the methane assays 99.2 per cent methane, .5 per cent ethane and .3 per cent nitrogen. Since no change in counter operation was observed from batch to batch, each cylinder of gas was not assayed.

$\Delta E/E$, the fractional full width at half-maximum of the spectral peak observed when monoenergetic radiation is admitted to the sensitive volume of the counter, is taken as a measure of the energy resolution of the instrument. This resolution has been measured using a 90 per cent argon - 10 per cent methane mixture for several external sources of quantum radiation and sources of internal conversion electrons mounted between thin films and inserted into the counter on the source probe. Table 1 contains a comparison of these experimental values of $\Delta E/E$ with the theoretical fractional full width at half maximum, $(\Delta E/E)_{theor.}$. If, for this purpose, a Gaussian distribution is assumed, $(\Delta E/E)_{theor.}$ is seen from Equation 11 to be given by

$$(\Delta E/E)_{theor.} = 2(2 \ln 2)^{1/2} \left[v_t/(m_A m_o)^2 \right]^{1/2} = 2.35 \, (2/m_o)^{1/2} = 3.32 \, (W/E)^{1/2} ; \quad (15)$$

[3] Available ready-mixed as "P-10" counter gas from The Matheson Company, Inc., East Rutherford, New Jersey.

where W, the energy required to form an original ion pair, is taken as 28 ev, and E is the energy of the incident radiation, all of which is assumed to be expended in the counter gas.

Table 1. Comparison of measured and calculated resolution of proportional counter

Source	Location	Type of radiation	Energy (kev)	$\frac{\Delta E}{E}$	$\left(\frac{\Delta E}{E}\right)_{theor.}$
Sb^{125}	external	x-ray	27.4	.086	.105
Ba^{133}	external	x-ray	30.8	.094	.100
Tb^{156}	external	x-ray	42.7	.086	.085
Te^{125m}	internal	conv. elect.	78	.094	.063
Te^{125m}	internal	conv. elect.	105	.085	.054
$Cs\text{-}Ba^{137}$	internal	conv. elect.	625	.034	.022

The modifications of Equation 11, which are discussed on pages 15 and 16, will serve to reduce the values of $(\Delta E/E)_{theor.}$ given in Table 1 by about 20 per cent at most.

These determinations of $\Delta E/E$ were run at gas pressures ranging from .6 atmospheres to 3 atmospheres of gas pressure. An external magnetic field was applied during the conversion electron measurements, but not during the x-ray measurements.

It can be seen from this tabulation that the disagreement between experiment and theory, while not great in the worst case, is appreciably less for quanta from external sources than for electrons from internal sources. Since the resolution for x-rays from an external source was

observed to remain unchanged by application of the external magnetic field or by insertion of the source probe (maintained at the correct potential), it seems unlikely that the differences in resolution are due to either magnetic field effects or electric field disturbances caused by the presence of the source rod and probe. More probably, the differences result from straggling and possible recombination along the tortuous paths of the more energetic electrons such as those from Cs-Ba^{137} and from energy losses in the source material and the support film in the case of softer electrons.

Moreover, any variations in anode diameter, power supply ripple or instability, amplifier noise or gain instability, gas impurities and analyzer drift will adversely affect the resolution to some extent regardless of the type of source; and none of these can be avoided completely at all times.

During the course of these preliminary evaluation studies, the thin source support film has in some instances been coated with a dilute insulin solution to reduce the surface tension of liquid sources, and normal precautions have been taken to prevent gross clumping and cratering of solid source residue; but no elaborate techniques were employed to reduce source thickness to an absolute minimum. Investigation with this counter of soft electrons, e.g., low energy beta spectra, could profitably employ either gaseous sources or solid sources prepared by vacuum evaporation methods similar to those devised by Pohm et al. (37).

In connection with the preparation of thin sources, the technique developed for the preparation of thin supports is of interest. One part of

clear Tygon paint[4] is diluted with two parts of Tygon thinner. One drop of this solution is dropped onto the surface of distilled water in a large tray; and after drying for a few seconds, the resultant film is lifted out with a submerged wire frame as shown in Figure 10, forming a double layer. Use of the slowly turning clock motor hoist avoids wrinkling and breakage of film, which frequently occurs when films are lifted by hand. Rectangular source support frames 1 inch long by 1/4 inch wide are formed from .005-inch diameter aluminum or nickel wire, dipped in very dilute shellac, placed on the film and cut out with the point of a hot needle. Double films with a mass of one to two micrograms per square centimeter can readily be produced in this manner. The source material is placed on the film, covered with another layer of film if desired; and the frame is attached to the end of the source rod, ready for insertion into the lock and subsequently into the counter. If a cover film is used, careful pre-evacuation of the source is recommended to avoid breakage in the source lock due to small entrapped air bubbles between the film layers.

The 625-kev K-conversion electrons from an internal source of Cs-Ba^{137} have been studied to ascertain the effect of the external magnetic field on the resolution of the electron line at a gas pressure much too low to prevent such energetic electrons from reaching the counter wall. A source of Cs-Ba^{137}, prepared by placing several pinpoint-sized drops of source solution on a thin Tygon film, allowing them to dry and covering with another film, was inserted into the counter on the internal source

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[4]Available from U. S. Stoneware, Akron 9, Ohio.

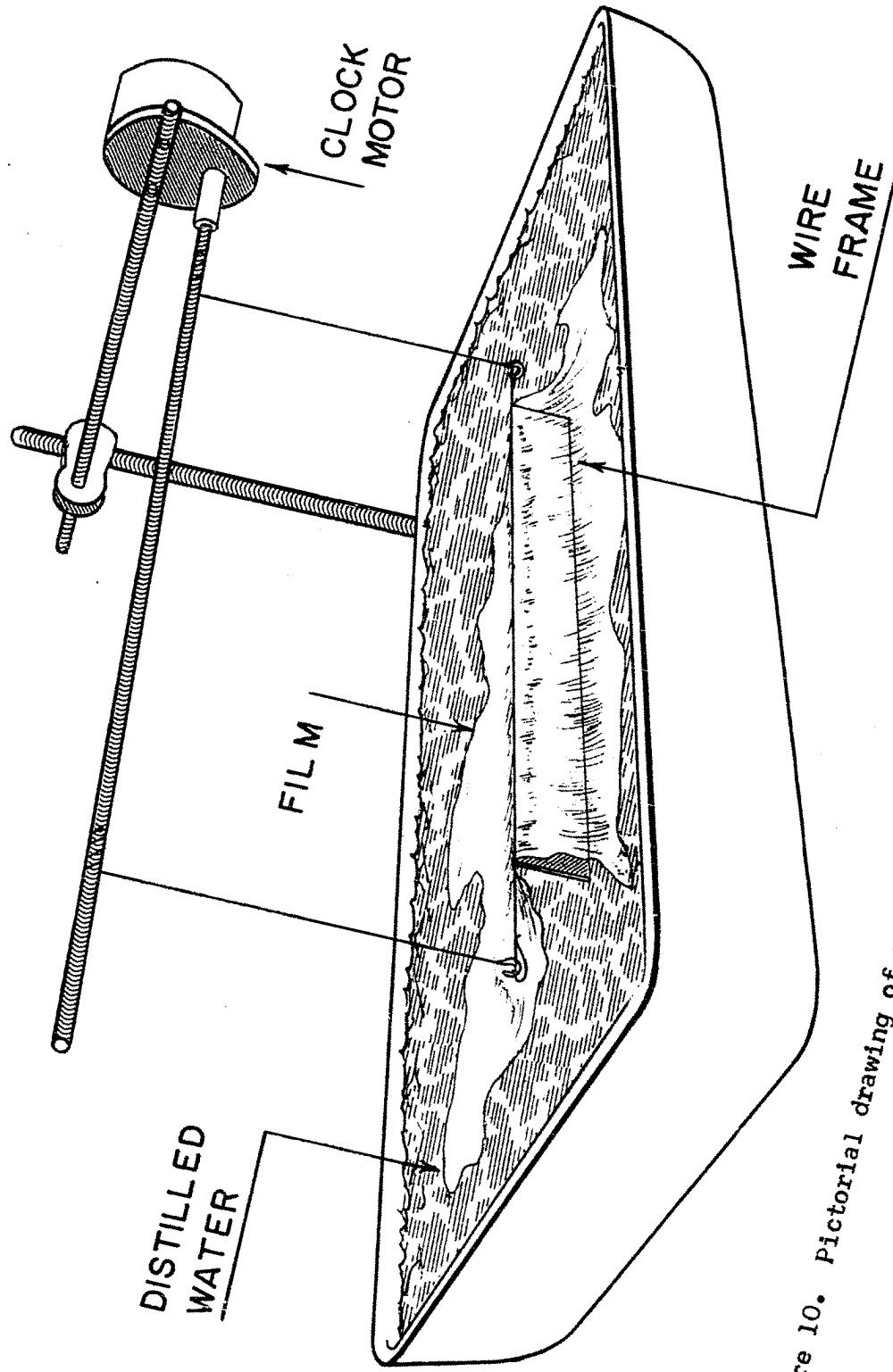

Figure 10. Pictorial drawing of apparatus for preparation of thin source support film

probe. With a gas mixture of 26 psi of argon and 4 psi of methane, and with the counter voltage adjusted to 4250 volts, the conversion electron spectrum was measured with the multi-channel photographic analyzer at various settings of the magnetic field, ranging from zero field to 2.5 kilogauss. Since conversion electrons which escape the counting volume will be recorded in lower analyzer channels, the ratio of the intensities at the conversion peak and at the valley between this peak and the continuous beta spectrum was taken as a convenient measure of the effectiveness of the field in constraining the electrons to the counter volume. The results are shown in Figure 11. The field of about 1.5 kilogauss required to constrain completely these 625 kev electrons is of interest in view of the fact that the diameter of curvature of such electrons in this field in a vacuum is only 2 inches, whereas the nearest counter wall is 4.5 inches from the probe. The electrons obviously do not move in uninterrupted circular paths, because of the large amount of scattering which takes place at this pressure; and at least half of the scattering events will throw electrons outside the cylindrical surface which would bound their orbits in a like magnetic field in vacuum. Figure 12 shows the internal conversion electron spectrum of Cs-Ba^{137} obtained at a magnetic field setting of 2.5 kilogauss.

In preparation for the coincidence measurements to be described in Chapter III, a careful measurement has been made of the time delay which occurs between the passage of ionizing radiation through the counter gas and the arrival of the output pulse at the analyzing instrument. An external source of Ba^{133}, which decays by K-capture, was placed against the thin (.016-inch) aluminum end of a reentrant cup set into one of the side

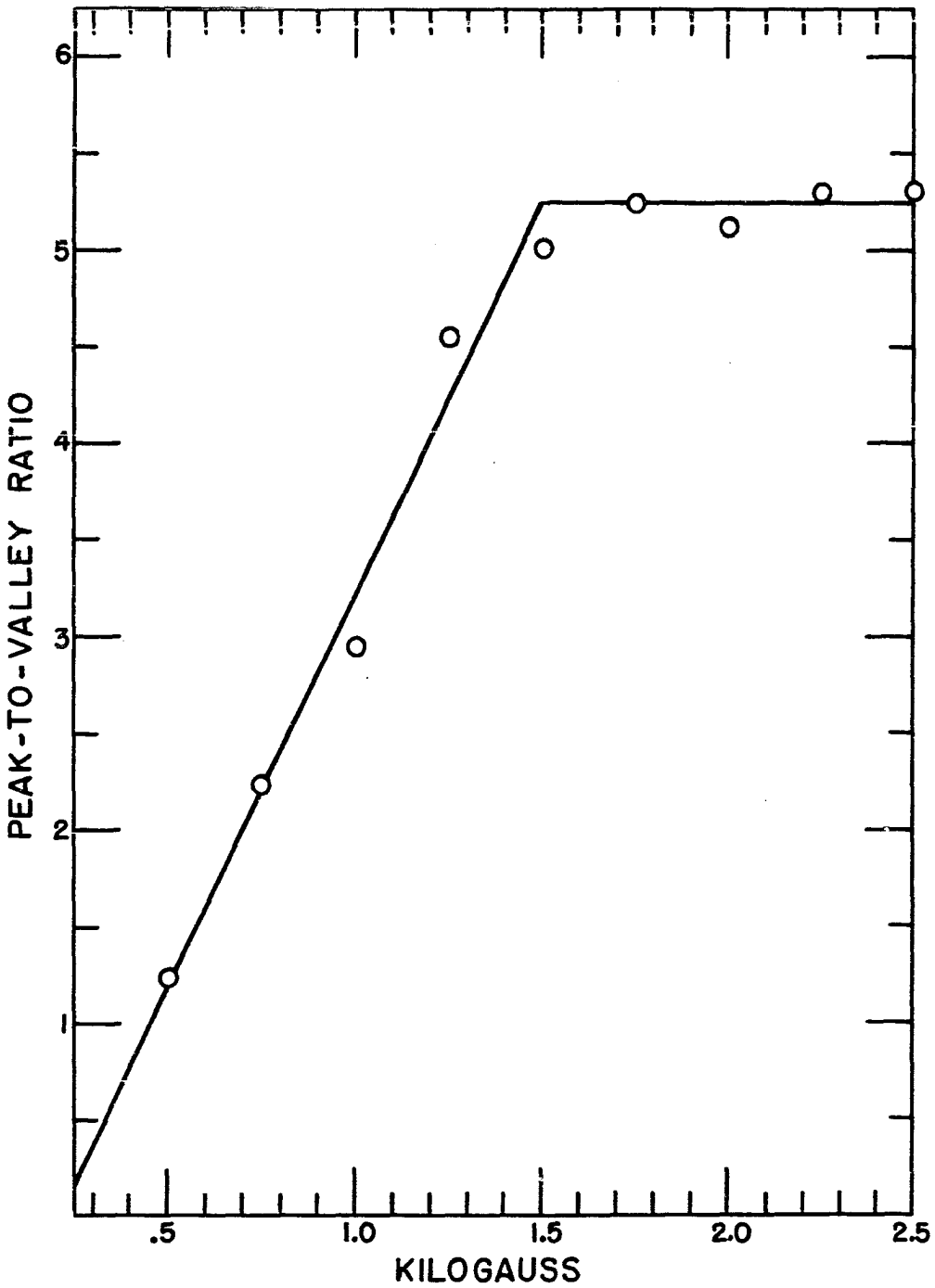

Figure 11. Effect of axial magnetic field on resolution of 625-kev electrons from internal source of Cs-Ba^{137}

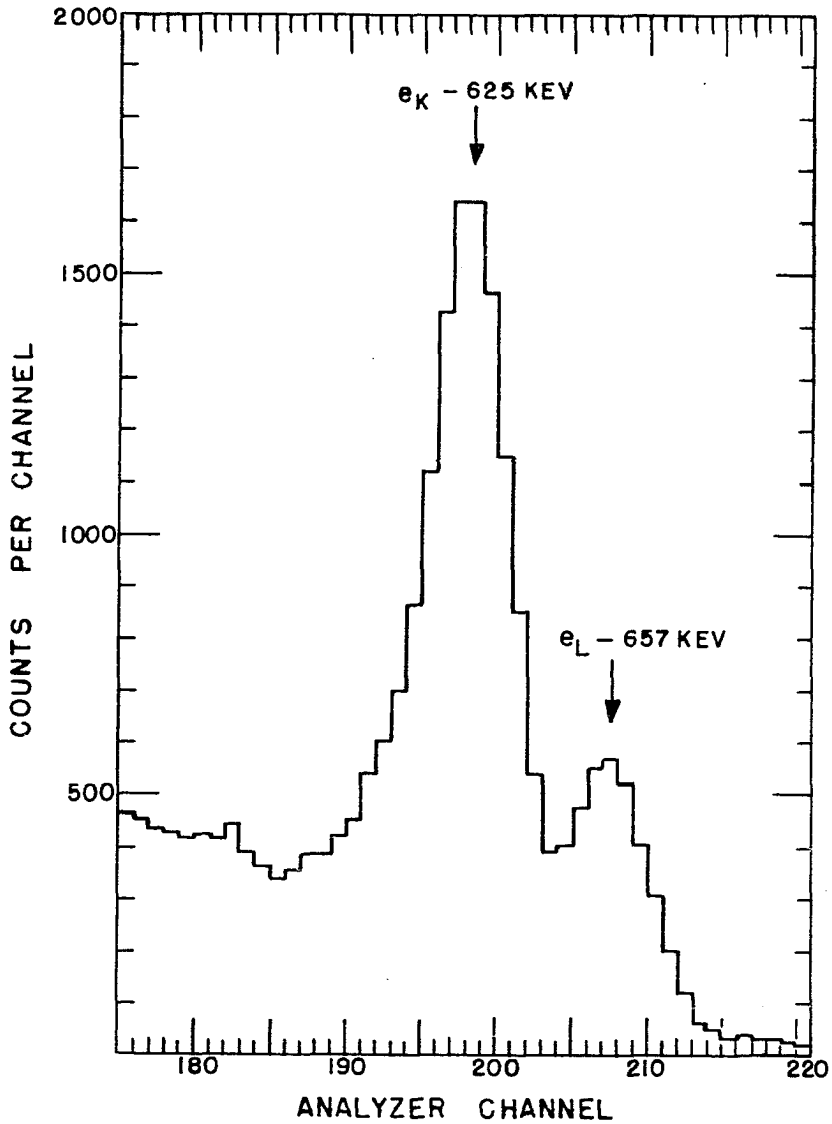

Figure 12. Proportional counter spectrum of conversion electrons from internal source of Cs-Ba^{137} in magnetic field of 2.5 kilogauss

ports of the counter so that nearly 2π steradians of sensitive volume could be seen by radiation from the source. A NaI(Tl) crystal mounted on an end-window photomultiplier tube (RCA Type 6655) was located against the other side of the source. Pulses from the photomultiplier were amplified and sent into an integral discriminator which triggered the calibrated linear sweep of a fast oscilloscope. The proportional counter pulses were amplified and passed through a single-channel differential analyzer which was adjusted to select only pulses corresponding to incident x-rays. The output of the single-channel analyzer was then applied to the Y-input of the oscilloscope, and the total delay introduced by the electron drift in the counter and the subsequent action of the single-channel analyzer was observed directly. For counter operation at 14 psi pressure of 90 per cent argon - 10 per cent methane gas mixture, and V_o = 3000 volts, a total delay of 9.7 microseconds was observed, more than half of which was indicated by later experiments to occur in the analyzer.

Utilizing the 27.4-kev x-rays from an external source of Sb^{125}, measurements have been made of output pulse height in the proportional region as a function of anode voltage for several gas pressures of a 90 per cent argon - 10 per cent methane mixture. The counter capacitance and the amplifier gain were determined by collateral experiments. Then, taking 28 ev as the energy required to form an ion pair and calculating from Equation 13 that 41 per cent of the total pulse amplitude was retained by the 10 microsecond clipping employed in the amplifier, pulse heights were converted to gas gain, which is tabulated in Table 2.

These data, in conjunction with Equation 4, should be helpful in

Table 2. Gas multiplication factor as a function of anode voltage for several pressures of a 90 per cent argon - 10 per cent methane gas mixture in a proportional counter with a = 6.35×10^{-3} cm and b = 15.24 cm.

V_o	Gas pressure (pounds per square inch)						
	3	7	12	14	15	20	25
1700	359						
1800	701						
1900	1390						
2000		89.4					
2100	6070	164					
2300		567					
2400			35.0				
2500		2110	57.0		15.5		
2600					28.5		
2700		7840	161	59.6	40.1		
2800					59.1		
2900			461	148	90.7		
3100			1420	404	238		
3300			4130	1100	635	75.1	
3400			6880				
3500				3220	1390	174	
3700				8130	3690	430	71.2
3800							105
3900						1062	159
4000							240
4100						2640	365
4200							562
4300						6000	860
4400							1310
4600							2980
4800							6150

estimating the gas amplification to be expected in other cylindrical counters using the same gas mixture.

B. Associated Instrumentation

1. Scintillation and Geiger-Muller counters

Most of the gamma-ray measurements have been made with a scintillation counter consisting of a Harshaw cylindrical thallium activated sodium iodide crystal 2 inches in diameter and 2 inches thick mounted with silicone grease directly on the 3-inch diameter flat face of a DuMont Type 6363 photomultiplier tube, which was selected for low internal noise and optimum resolution. A cathode follower of orthodox design, employing one 6AK5 tube, has proven adequate to drive the necessary length of cable between the counter and the 256-channel analyzer. In all spectral measurements, pulses from the scintillation counter were amplified by the A-61 linear amplifier which comprises part of the 256-channel analyzer.

Efforts to detect electrons by scintillation methods employed an anthracene crystal 1.5 inches in diameter and .25 inches thick, mounted with silicone grease directly on the face of a selected RCA Type 6655 photomultiplier and covered with a layer of aluminum leaf which served as a light reflector. This leaf has a mass of only .12 milligrams per square centimeter. Pulses from this phototube were fed through a dynamic cathode follower into the A-61 linear amplifier in the 256-channel analyzer as in the previous case.

The G-M counter used for long half-life observations was an end-window Tracerlab Type TGC-3NA, whose mica window has a density of 1.9 mil-

ligrams per square centimeter. This tube was mounted in a lead counter pig and operated on its experimentally determined plateau in conjunction with a conventional power supply and scaler.

2. Pulse height analyzers

After amplification, the output pulses from the proportional counter or any of the scintillation counters may be subjected to amplitude analysis by one of four available methods:

a. The integral pulse height discriminator which is accessory to the Model 101 linear amplifier, followed by a scaler and, if desired, an automatic printer.

b. A single-channel differential pulse height analyzer followed by a scaler.

c. A "dot-scope", in which a short section of the flat top of a properly shaped pulse is intensified to a small (.005-inch diameter) dot on the face of a cathode ray tube. These dots, each of whose distance from a fixed base line is proportional to the pulse amplitude, are photographed on continuously moving film which, after development, is optically scanned by a 200-channel photoelectric pulse height analyzer (36).

d. A Radiation Counter Laboratories Mark 20 Model 2603 (Argonne Type) 256-channel pulse height analyzer based on the design of Schumann and McMahon (26). As mentioned previously, this analyzer includes a linear amplifier, designated Type A-61, which may be used instead of a separate amplifier if desired. In addition, the analyzer design contains provisions for acceptance of either prompt or delayed coincidences, thus obviating the need for auxiliary coincidence circuits.

III. RADIATION FROM Tb^{156}

A. Irradiation of Source Material

Three groups of terbium samples have been irradiated for varying lengths of time in the external 64-Mev bremsstrahlung beam of the Iowa State College electron synchrotron. The nature of the sample, the duration of the irradiation and the uses for which the various samples were intended are indicated in Table 3.

Table 3. Terbium irradiation data

Run no.	Duration (hours)	Form of sample	Subsequent use
1	16	5.45 g Tb metal slug 300 mg Tb metal slivers	First survey of γ spectrum G-M half-life determination
2	136	2.93 g Tb_4O_7 230 mg Tb_4O_7	Measurement of γ spectrum, γ-x-ray coincidences G-M half-life determination
3	5	882 mg Tb metal foil 433 mg Tb metal foil	Decay of selected γ-rays G-M half-life determination

B. Measurement of Spectra and Decay Rates

Run 1 was intended as an exploratory irradiation to confirm the presence of the anticipated $Tb^{159}(\gamma,3n)Tb^{156}$ reaction and to allow a preliminary examination of the intensity, decay rate and energy range of the induced activity. After irradiation, a sample of terbium metal consisting of several small slivers with a total weight of 300 milligrams, was placed on the top shelf in a G-M counter pig and the total activity

was recorded at 15-minute intervals with a Streeter-Amet automatic printer for a period of 25 days after the end of the irradiation. The results are shown in Figure 13. A few flags are shown to indicate errors in the experimental points, and a $T_{1/2}$ = 5.6-day curve has been included to indicate the degree of agreement with one of the previously reported (21) half-lives of Tb^{156} activity.

An examination of electromagnetic radiation from the 5.45-gram slug of terbium metal revealed a number of gamma-ray lines with energies in rough agreement with those reported by Handley and Lyon (19) but no activity in the vicinity of 90 kev which could be seen to decay with a half-life approaching 5 hours as reported by previous investigators (18, 19,20,21). Accordingly, two other irradiations were scheduled and made: a long bombardment to provide a high intensity of 5.6-day activity and a short exposure to enhance any 5.5-hour activity which might be masked by the long-lived components produced during a long irradiation. These are designated as irradiations 2 and 3 in Table 3.

After irradiation 2, the 230-milligram sample of Tb_4O_7 was placed in the G-M counter pig with the intention of repeating the total activity half-life experiment, but unfortunately the printing circuit failed and no useful data were obtained.

The large oxide sample, comprising 2.93 grams of Tb_4O_7 in the form of a fine powder, was suspended in distilled water and then filtered by aspiration, causing the oxide to settle out on filter paper over a circular region 6 centimeters in diameter, with a density of approximately 100 milligrams per square centimeter. After drying, the source was

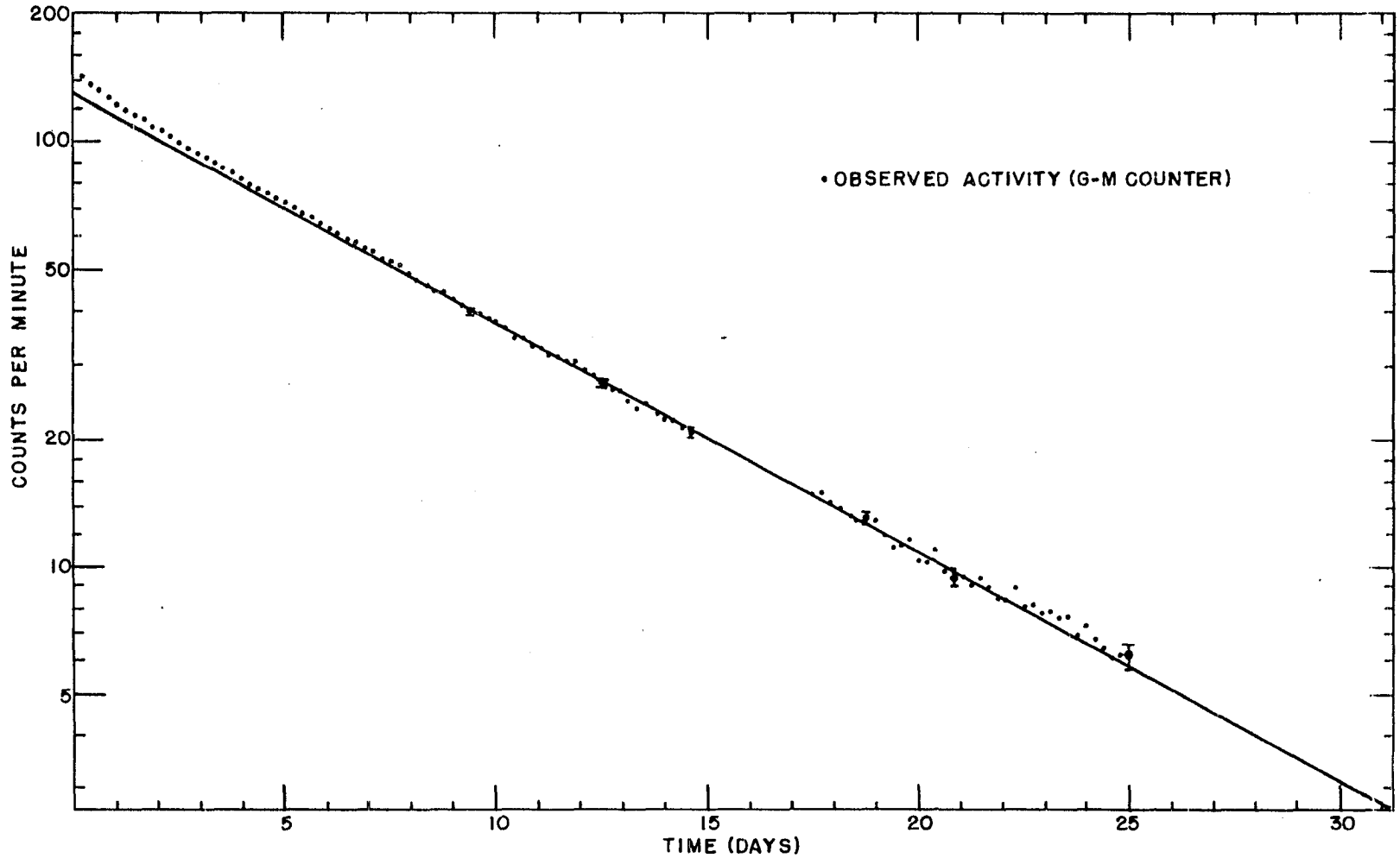

Figure 13. G-M counter measurement of decay of total activity induced in terbium metal by a 16-hour irradiation, compared with a curve of $T_{1/2}$ = 5.6 days

covered on both sides with a layer of cellophane tape.

This source was placed 2 inches away from the 2-inch diameter NaI(Tl) crystal described in Chapter II, and the gamma-ray spectrum shown in Figure 14 was obtained with the help of the 256-channel analyzer. Calibration of these gamma-ray energies against known lines in Co^{60} and identifiable lines reported by Mihelich et al. (21) is shown in Figure 15. A semi-logarithmic plot was necessary because of the wide range of intensities present.

Calibration was accomplished in two steps. First, a spectrum was recorded for a composite source of Tb^{156} and Co^{60}. In this spectrum, peaks were seen at approximately 200 kev and 350 kev, so these were tentatively assumed to be the transitions reported by Mihelich et al. (21) at 199.4 kev and 356.6 kev. The two Co^{60} lines at 1.17 Mev and 1.33 Mev were also clearly distinguishable. From the straight line drawn through these four points, energies were assigned the three transitions indicated by circles in Figure 15, thus yielding a total of five plainly discernable peaks of known energy in the Tb^{156} spectrum. Another much longer analysis was then made of the spectrum of Tb^{156} alone, and the remaining visible peaks were assigned energies on the basis of a straight line drawn through these five fixed points. The energies shown for the six lowest-energy lines in Figure 14 (designated by squares in Figure 15) are due to Mihelich et al. (21), whose precision exceeds that of the present experiment. However, in the spectrum shown in Figure 14, there is an ambiguity of no more than one analyzer channel in the location of most of the peaks; and in Figure 15, the largest deviation of a calibration point from the straight line

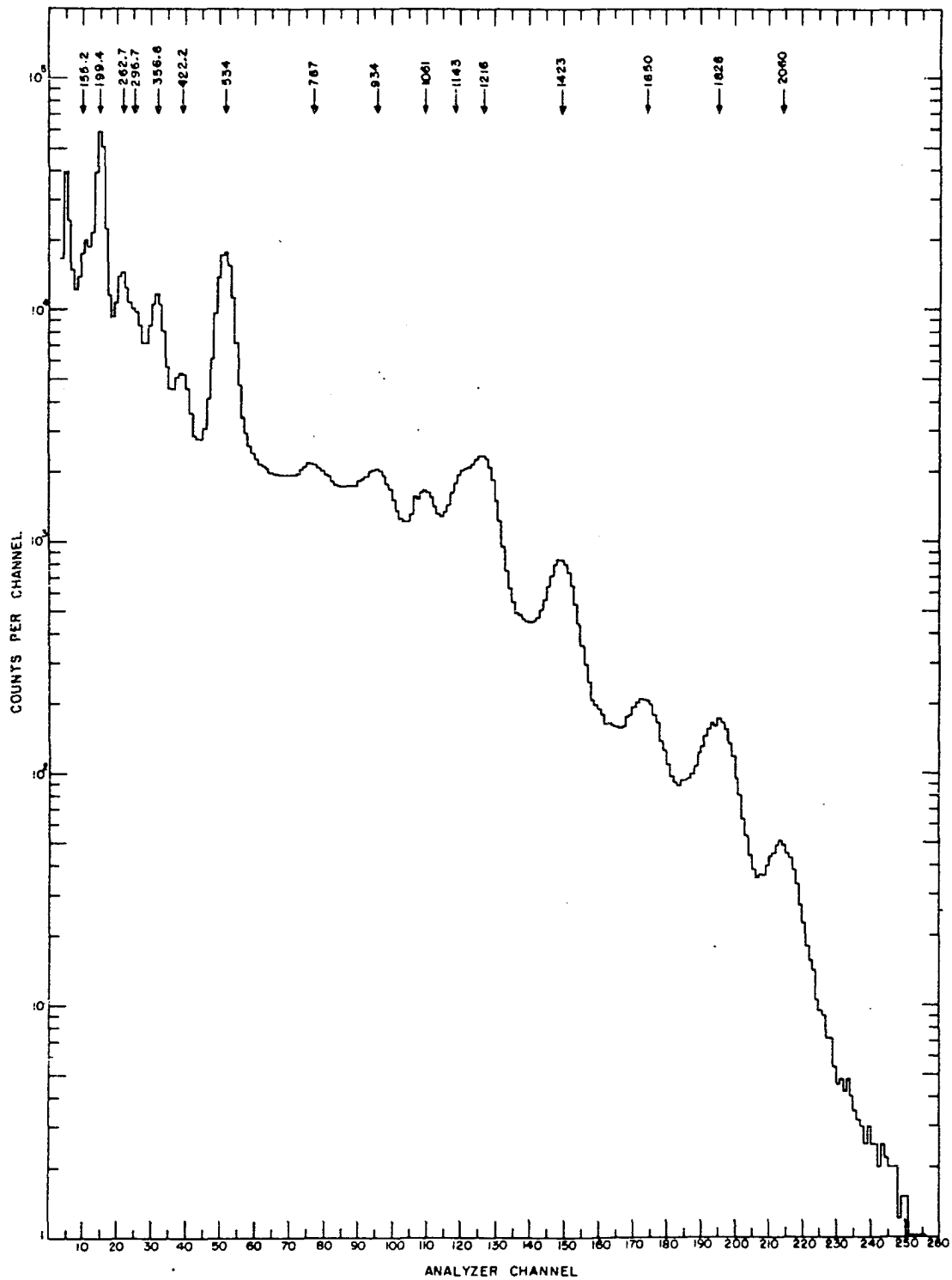

Figure 14. Scintillation spectrum of electromagnetic radiation from Tb_{47}^{Q} sample following 136-hour irradiation; energies are in kev

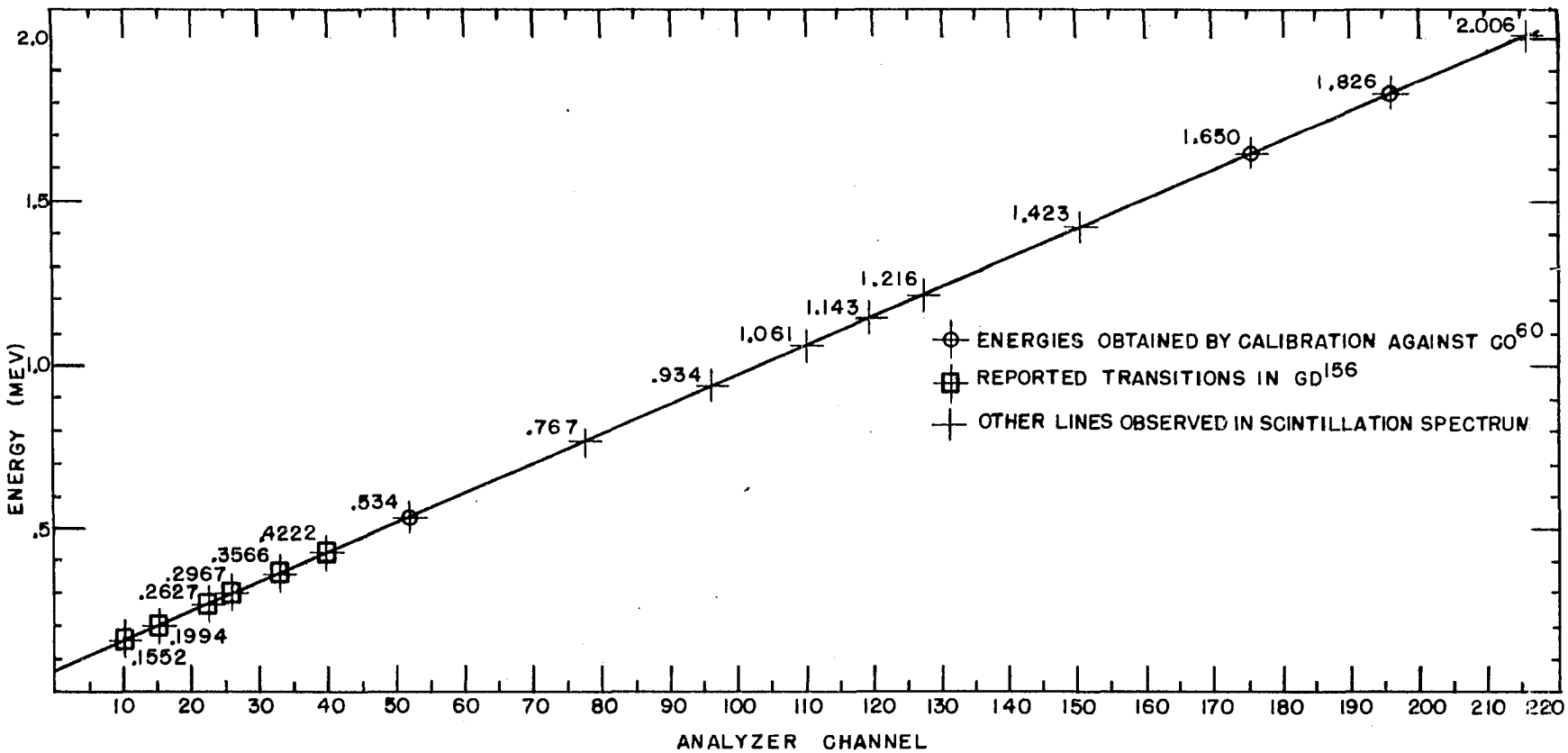

Figure 15. Energy calibration of transitions observed in the scintillation spectrum of Tb^{156} as shown in Figure 14

is approximately 5 kev.

After this gamma-ray singles spectrum was recorded, the source was placed in the aluminum cup in one of the side ports of the large proportional counter. The proportional counter was filled to 14 psi with a 90 per cent argon - 10 per cent methane gas mixture, a voltage of 3000 volts was applied to the anode, and the amplifier gain was adjusted so that a photon with an energy of 50 kev would produce an output pulse of sufficient amplitude to appear at about the middle of the 256-channel analyzer display range. The result of a 33.3-minute run under these conditions is shown by the solid histogram in Figure 16, in which the major peak is at 42.7 kev, as expected for x-rays from gadolinium. The resolution of the proportional counter for radiation in this energy region is crucial to the subsequent coincidence study. The dotted curve in Figure 16 represents the predicted location of the Dy x-ray; and it is apparent from the resolution of the Gd x-ray peak that a Dy x-ray would be visible if present in any appreciable quantity and that little or none was present. The proportional counter output was then sent into a single-channel analyzer whose differential discriminator window was centered on the Gd peak and set at a width equal to the full width of this peak at half maximum. This setting would pass three-fourths of all Gd K x-ray pulses and only one-eighth of any Dy K x-rays which might be present. Each output pulse from the single-channel analyzer triggered a simple univibrator gating circuit (Figure 17), which in turn energized the coincidence input of the 256-channel analyzer for an interval of three microseconds. During this interval any pulses arriving from the photomulti-

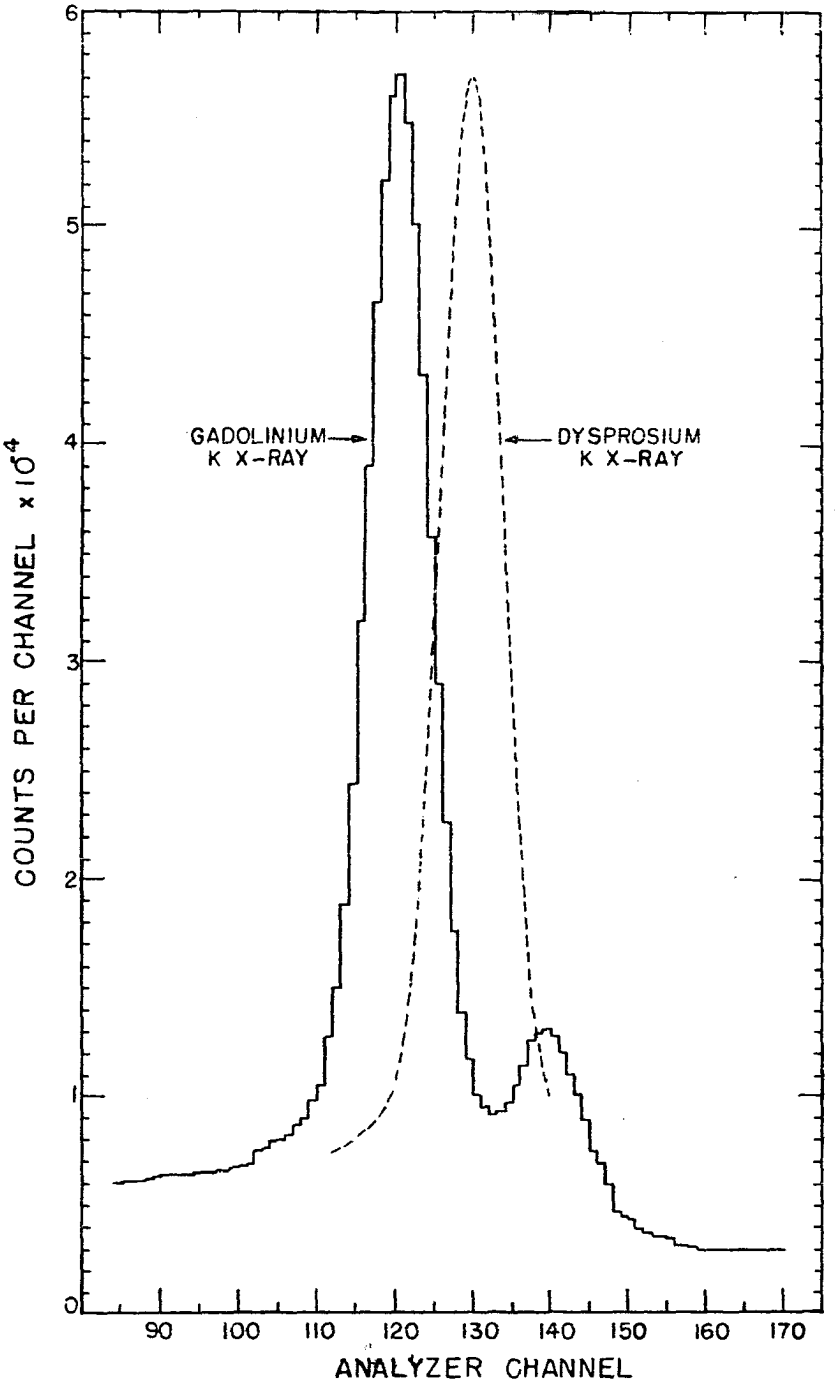

Figure 16. Proportional counter spectrum of x-radiation from external Tb_4O_7 sample

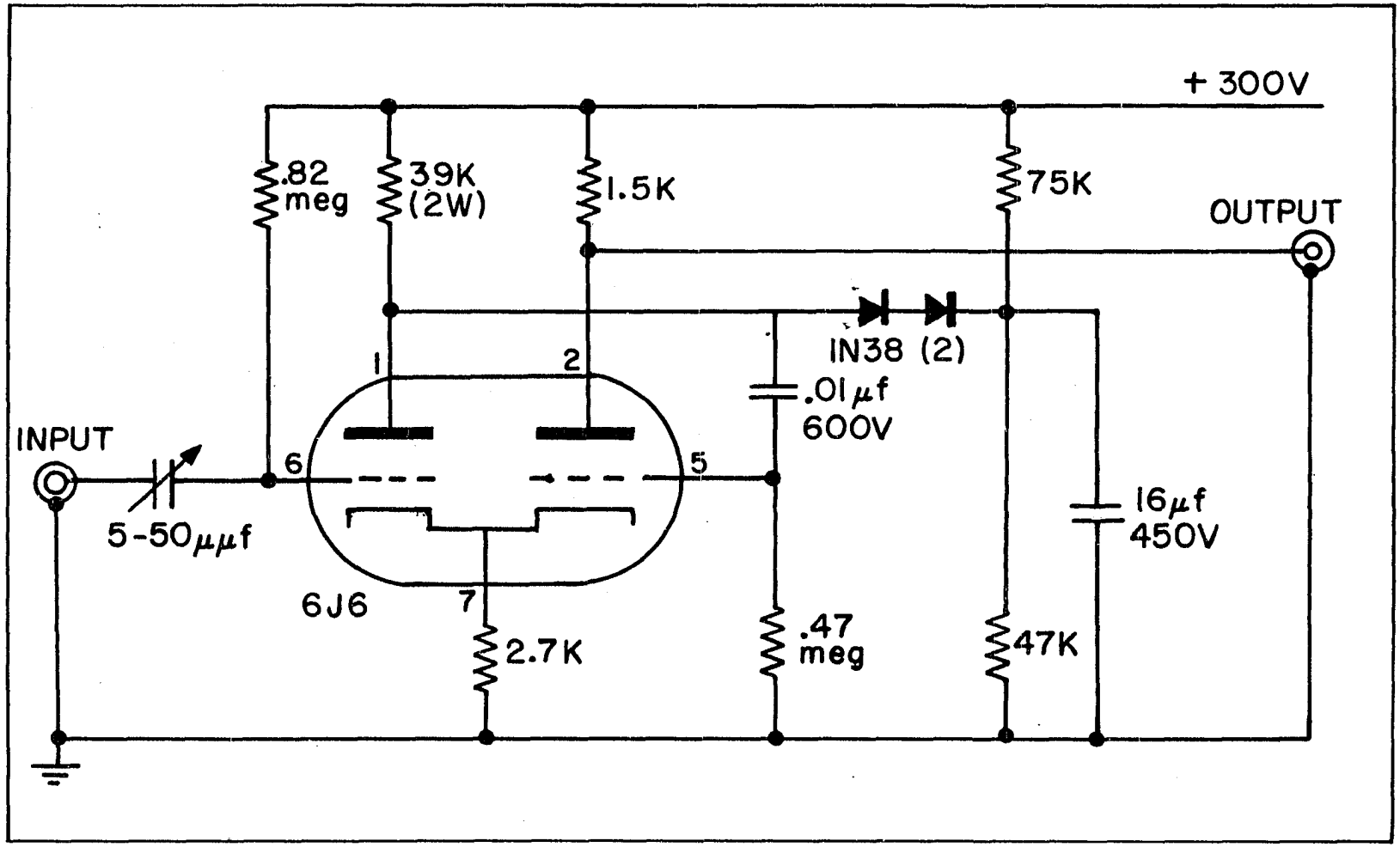

Figure 17. Univibrator gating circuit

plier tube were accepted and analyzed. The length of the time gate was determined by visual inspection of the time "jitter" of pulses from the proportional counter which indicated that at least half of the truly coincidental pulses arrived within a 3-microsecond interval. A delay line consisting of 14.5 feet of HH 2500 cable, which has a delay of .6 microseconds per foot, was interposed between the photomultiplier and the analyzer. This delay of 8.7 microseconds, when added to the 1-microsecond delay which was found to occur in the 256-channel analyzer coincidence circuit, was required to compensate for the 9.7 microsecond delay in the proportional counter and the single-channel analyzer which was mentioned in Chapter II. When properly terminated, this delay line caused no observable distortion in the spectrum being analyzed.

The gamma-ray spectrum thus obtained in coincidence with the Gd x-rays is shown in Figure 18. Although the resolution has suffered somewhat as a result of the relatively low coincidence counting rate and the correspondingly severe demand upon the long-term stability characteristics of the electronic circuits involved, most of the peaks visible in the γ-ray spectrum of Figure 14 are plainly discernable here. Exceptions are the very low intensity peak at 2.06 Mev, the peak at 155.2 kev and the adjacent peaks at 262.7 and 296.7 kev. The intensity of the 2.06-Mev transition was so low that no attempt was made to include it in the coincidence run, since this would have required further compression of the energy scale and an increase in counting time by perhaps a factor of two. The 155.2-kev peak is probably obscured by the line at 199.4 kev, and it appears quite likely that the peaks at 262.7 and 296.7 are present but

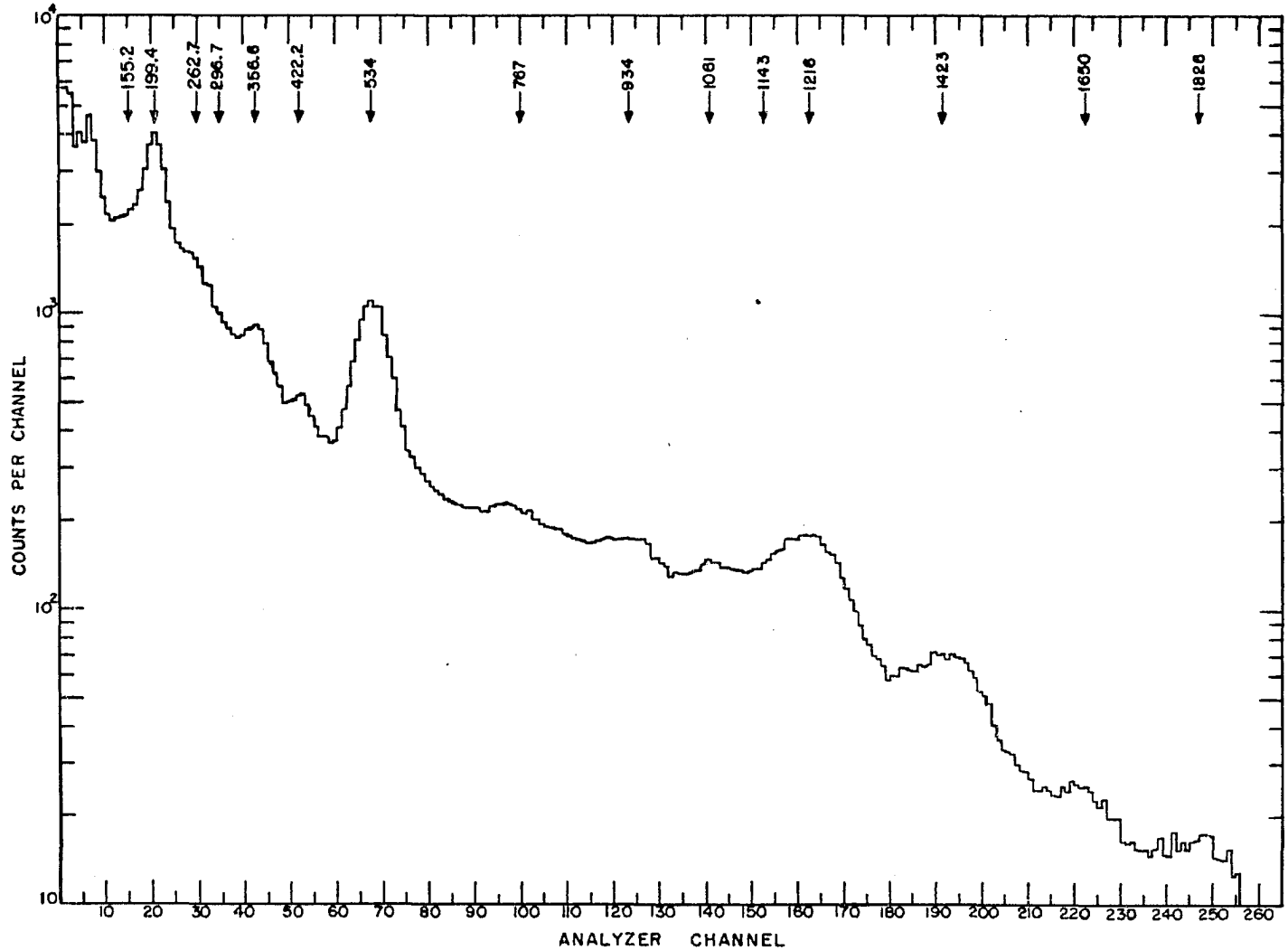

Figure 18. Scintillation spectrum of electromagnetic radiation from Tb^{156} in coincidence with x-rays from Gd

unresolved.

Irradiation run 3 was designed to display to best advantage any short-lived activity which might have been masked by longer-lived components produced during the longer previous bombardments.

The results of a G-M measurement of total activity in the 433-milligram sample of terbium foil for 150 hours after completion of irradiation are given in Figure 19. Some activity with a half-life considerably shorter than 5.6 days is clearly present. Subtraction of an assumed 5.6-day component leaves residual activity which fits a curve of $T_{1/2}$ = 12.6 hours quite well at the lower end, and a further subtraction of this activity yields a third component which is not in disagreement with the assumption of a 5.5-hour half-life.

While this G-M run was in progress, the gamma-ray spectrum from the simultaneously irradiated 882-milligram sample of terbium metal foil was examined at frequent intervals over a period of 127 hours with the scintillation spectrometer and the 256-channel analyzer. These successive records of the gamma-ray spectrum revealed a general tendency for the activity to decay with a 5.6-day half-life, but three energy regions seemed deserving of special scrutiny:

a. A peak at 124 kev which exhibited a decay rate greater than 5.5 hours and less than 5.6 days.

b. The region around 90 kev, which should include the 88.4 kev isomeric transition with $T_{1/2}$ = 5.5 hours and the 89.10 kev transition in Gd^{156} reported by Mihelich *et al.* (21).

c. The x-ray region in the vicinity of 43 kev.

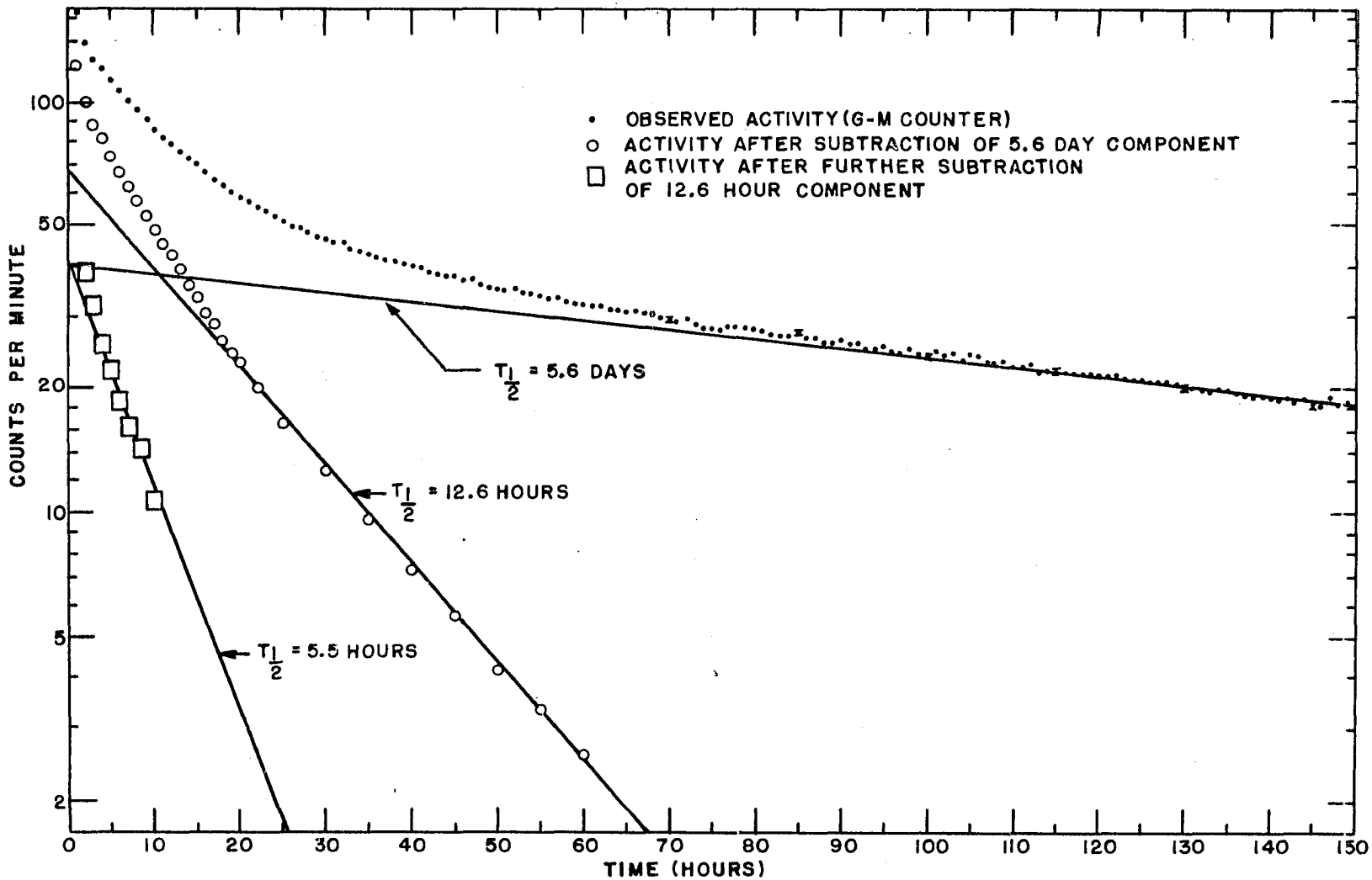

Figure 19. G-M measurement of decay of total activity induced in terbium metal by a 5-hour irradiation

The decay of the 124-kev peak is shown in Figure 20. Subtraction of the 5.6-day activity upon which this peak rides leaves a component with a half-life of 12.6 hours.

Figure 21 shows the decay of the x-ray peak in the vicinity of 43 kev. After subtraction of the underlying 5.6-day background, it appears that the remaining activities may reasonably be attributed to 12.6-hour and 5.5-hour activities as indicated; although the errors inherent in a double subtraction operation of this sort are so large and the amount of 5.5-hour activity thus revealed is so small that these data serve mainly to place an upper limit on the intensity of this component.

This same division into three components of 5.6 days, 12.6 hours and 5.5 hours is seen to fit the decay of the 90-kev peak shown in Figure 22. Inspection of the original data as recorded by the 256-channel analyzer indicates that most, if not all, of the 12.6-hour activity seen here in due to the tail of the previously mentioned peak at 124 kev.

In addition to these measurements of total activity and selected regions of the gamma-ray spectrum, a search has been made for the beta groups which have been reported by several investigators (18,19,24) to be associated with the decay of Tb^{156}. After irradiation 1, a small sliver (total weight 24.4 milligrams) was cut from the 5.45-gram slug of terbium metal and rolled into a foil with an area of approximately 1 square centimeter. Although not a particularly thin source, such a foil was judged adequate for the purpose of this research; since the softest beta group seen by Handley and Lyon (19) was reported to have a maximum energy of 140 kev.

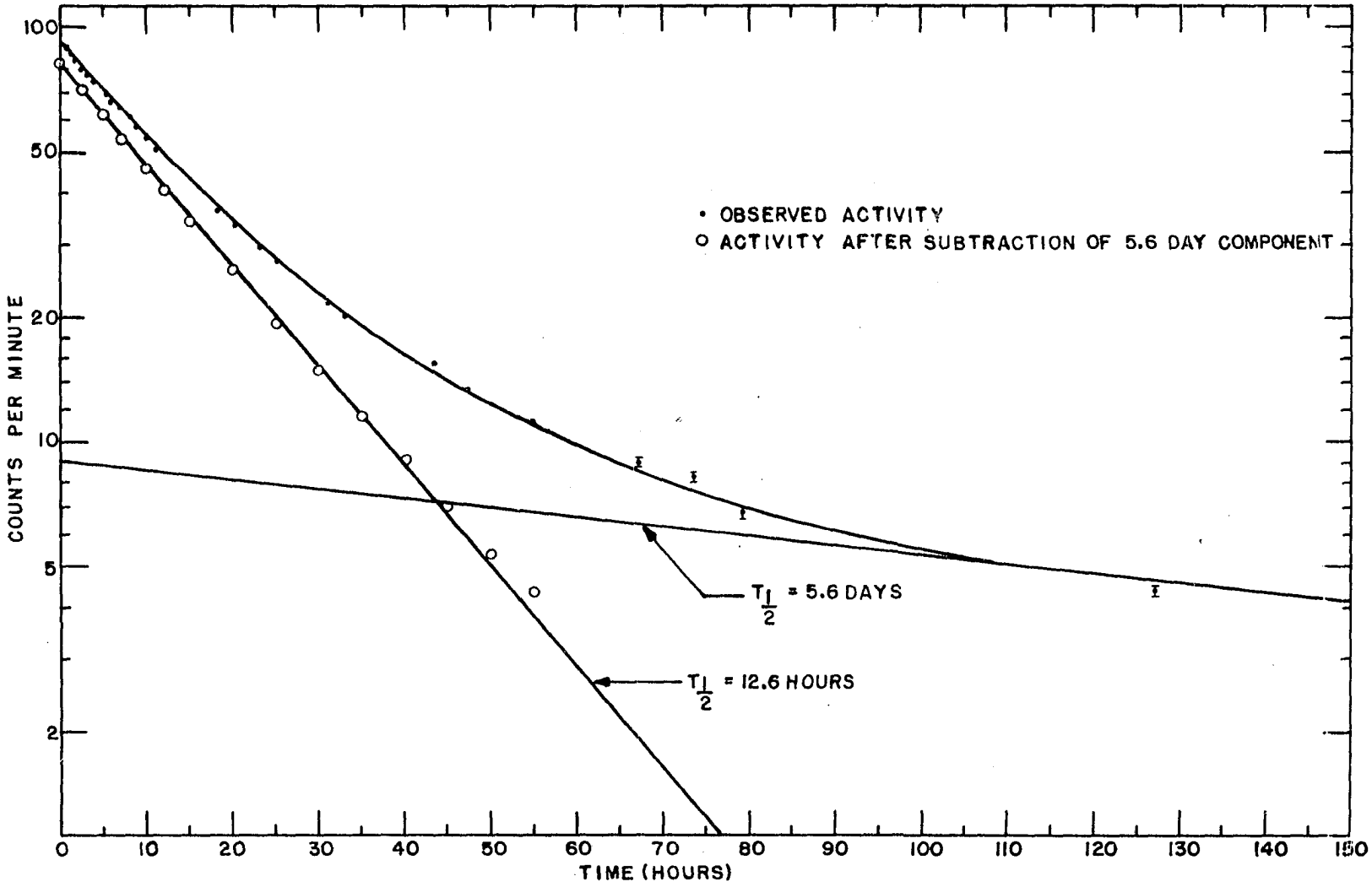

Figure 20. Scintillation counter measurement of decay of 124-kev peak observed in electromagnetic radiation spectrum of terbium metal after a 5-hour irradiation

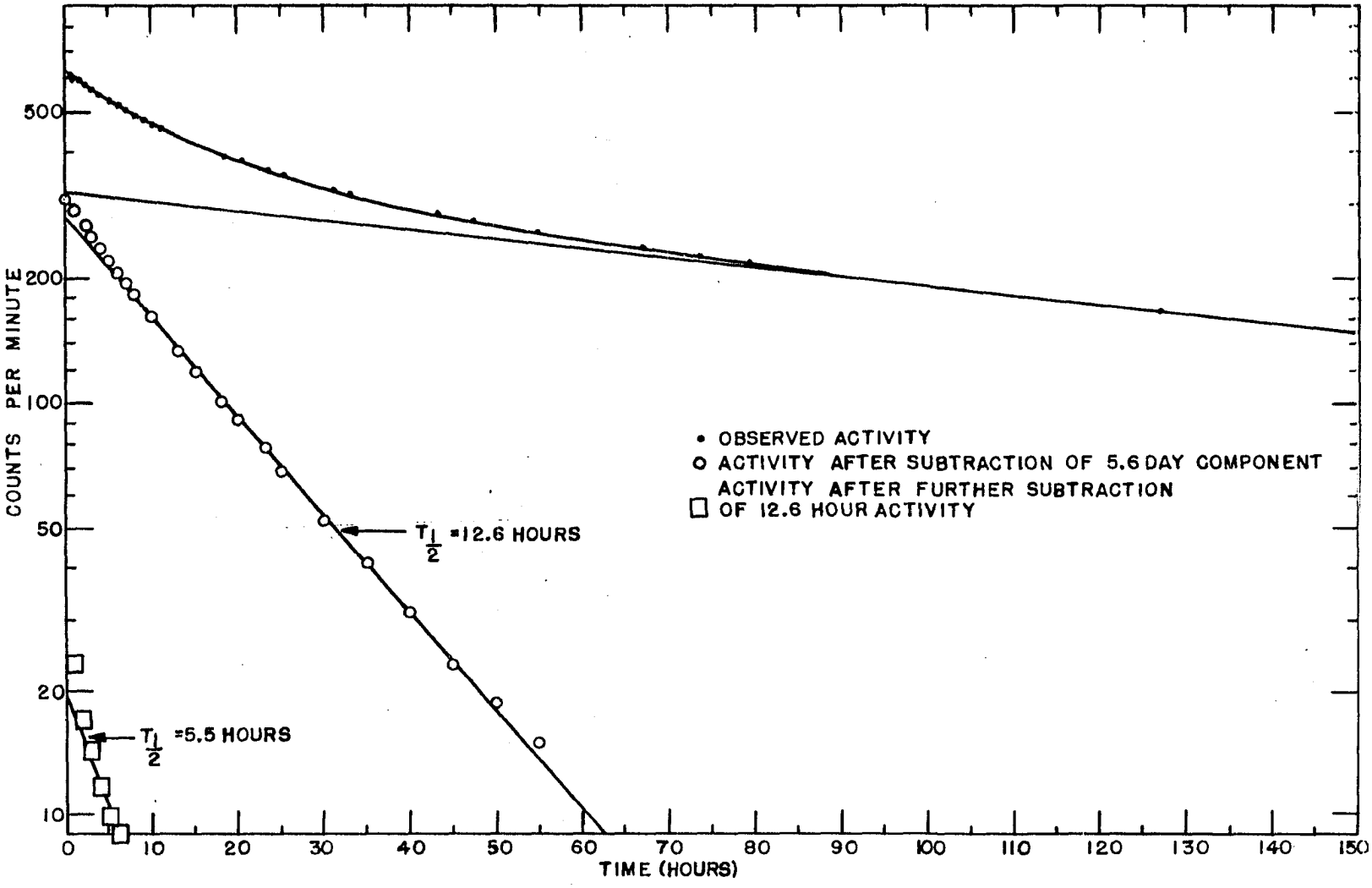

Figure 21. Scintillation counter measurement of decay of x-radiation observed at approximately 43 kev in electromagnetic radiation spectrum of terbium metal after a 5-hour irradiation

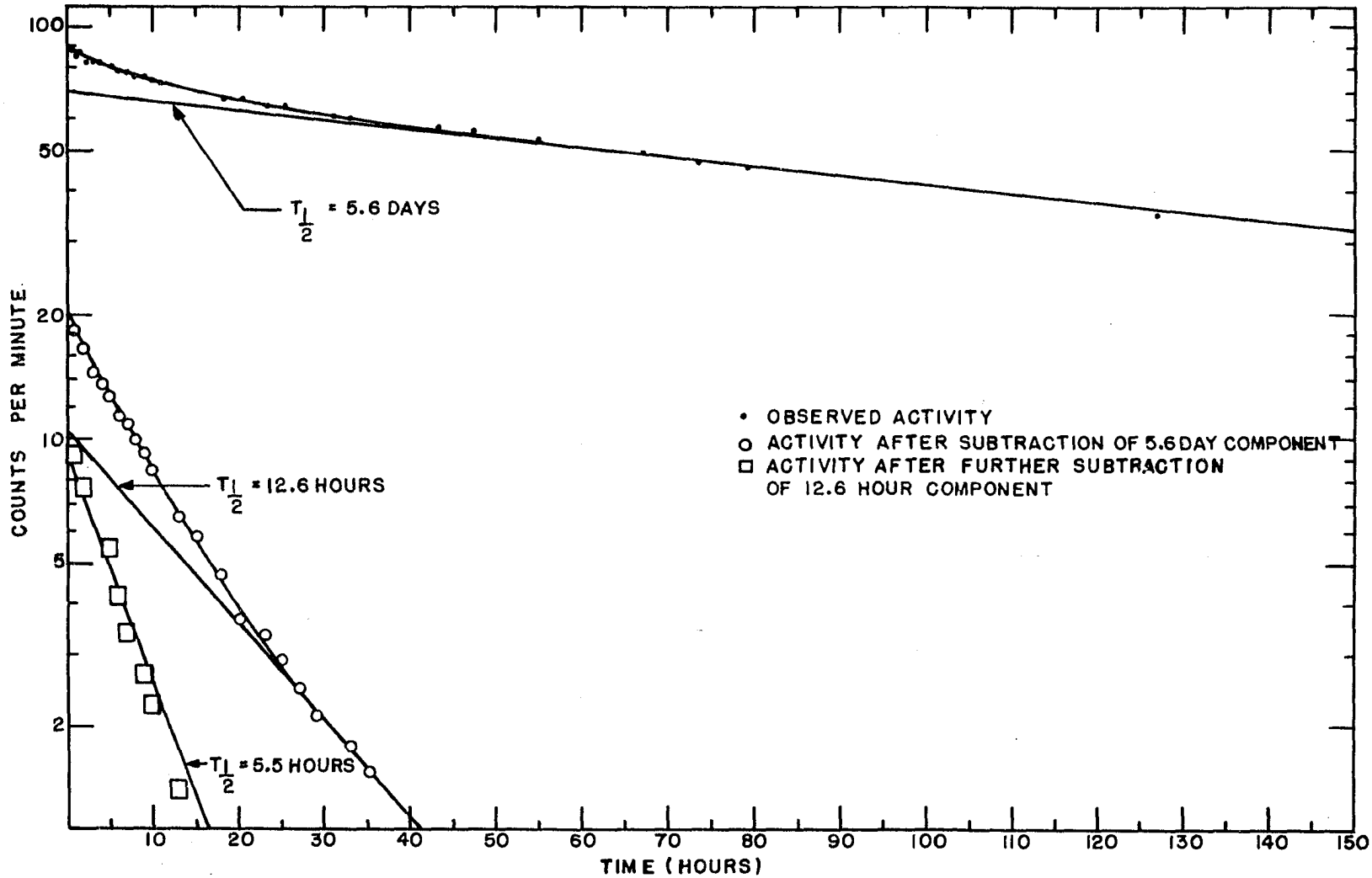

Figure 22. Scintillation counter measurement of decay of 90-kev peak observed in electromagnetic radiation spectrum of terbium metal after a 5-hour irradiation

When this foil was inserted into the proportional counter on the internal source probe, the resultant counting rate was measurably higher than background; but a spectral analysis revealed no clear evidence for the existence of any beta groups. A similar spectral shape was obtained when radiation from an external terbium metal source was admitted to the counter through a 1/8-inch thick beryllium window, which should have stopped any electrons emanating from the Tb^{156} with energies below about 1.3 Mev.

Subsequently, radiation from the same terbium foil was examined with a scintillation spectrometer employing a 1/4-inch thick anthracene crystal covered only with aluminum leaf which weighs .12 milligrams per square centimeter. With the help of the 256-channel pulse height analyzer, a spectrum was obtained which, although somewhat distorted, suggested the possible presence of two beta groups with end points at about 200 kev and 600 kev as reported by Handley and Lyon. An aluminum absorber .030 inches thick (sufficient to stop electrons with energies below approximately 600 kev) was then interposed between the source and the crystal. The counting rate was decreased, but the shape of the spectrum was found to be substantially unchanged. Consequently, it seems highly unlikely that the observed spectrum was that of beta-rays emitted from the terbium source.

IV. SUMMARY AND DISCUSSION

Data have been presented which give ample evidence of the utility of the large proportional counter spectrometer in the measurement of the energies of electrons and low-energy quanta from radioactive materials. The resolution of this instrument for electrons with energies in excess of 500 kev approaches that of some magnetic beta-ray spectrometers; and its much poorer resolution at low energies is still clearly superior to that of its chief competitor in this region, the scintillation spectrometer, for either electrons or quanta. The adaptability of the counter to coincidence measurements and the advantages of its relatively good resolution for low-energy quanta have been demonstrated in its application to the investigation of the radiation from Tb^{156}.

Future studies of the counter's characteristics might profitably include a more detailed examination of the dependence of its performance upon changes in gas composition, anode size and cathode diameter, which latter parameter may readily be decreased by the insertion of cylindrical liners. Full exploitation of its potential usefulness as an analytical tool also requires improvement in the preparation of thin internal sources, experimentation with the use of gaseous sources and further evaluation of its operating properties at higher gas pressures and correspondingly higher voltages than those used to date.

However, even at its present stage of development, the large proportional counter constitutes a valuable complement to the other techniques currently available for the measurement of nuclear energy spectra.

Several conclusions may be drawn regarding the radiation observed

after the irradiation of terbium by bremsstrahlung with a maximum energy of 64 Mev. In this radiation, 18 transitions have been detected with energies between 90 kev and 2060 kev, 16 of which are displayed in Figure 14. When the amplifier gain was increased sufficiently to expand the energy scale of this spectrum by a factor of four, the lowest eight peaks were found to agree (within the limits of the resolution of the scintillation spectrometer) with the eight transitions reported by Mihelich, et al. (21) to occur in Gd^{156} following the 5.6-day decay of Tb^{156}. No other transitions were observed in this region. Mihelich has given the energies of these lines as 89.10, 111.9, 155.2, 199.4, 262.7, 296.7, 356.6 and 422.2 kev. The present experiment cannot improve on these energy allocations, but their presence in the spectrum of the radiation studied herein, agreement of other observed transitions at 534, 767, 1216, 1423, 1826 and 2060 kev with those reported by Handley and Lyon (19) and the decay of this activity with a 5.6-day half-life form the main basis for the assignment of the activity presently under consideration to Tb^{156}. As mentioned in the previous chapter, 199.4 kev and 356.6 kev have been used as fixed points in the calibration of the observed energy spectrum.

Coincidences have been found between all of the observed transitions (except the one at 2060 kev) and x-rays from Gd, implying decay of Tb^{156} by orbital electron capture. Due to its low intensity, no evidence has been obtained for or against coincidences between the 2060-kev transition and these x-rays.

No evidence of beta-decay has been found. It is suggested that the spectrum obtained with an anthracene crystal during the search for beta

activity is probably due to photoelectrons and Compton electrons produced in the anthracene by the numerous gamma-rays in the Tb^{156} spectrum. Furthermore, if Tb^{156} decays to Dy^{156} by more than one beta group, it might be anticipated that some internal conversion would accompany de-excitation of the daughter nuclide; but no Dy x-rays are apparent in the proportional counter spectrum shown in Figure 16.

No evidence has been observed of annihilation radiation which would point to the presence of positrons.

One puzzling aspect of the present data is the paucity of 5.5-hour activity which has been reported so frequently by previous investigators (18,19,20,21,24,25). Handley and Lyon (19) have reported a ratio of 5.5-hour to 5.2-day activity of 80 to 1 on the basis of G-M counting; yet the data shown in Figure 19, after correction for time of irradiation and decay, indicate a ratio of about 1 to 18. Similarly, Mihelich et al. (20,21) report convincing evidence for the existence of an 88.4-kev isomeric transition in Tb^{156}, whereas the data shown in Figure 22 (again after correction) indicate that less than 1 per cent of the electromagnetic radiation emitted with an energy around 90 kev has a half-life near 5.5 hours. If the isomeric transition does occur and is very highly converted, one might expect a larger 5.5-hour contribution to the above mentioned G-M measurement of total activity.

It is difficult to appraise the findings of Dillman, et al. (24,25), since it seems likely that irradiation by 240-Mev bremsstrahlung may have produced other radionuclides than the one under consideration here. However, several of the gamma-ray energies reported by them are in agreement

with transitions observed in this investigation.

The origin of the 124-kev electromagnetic radiation with a 12.6-hour half-life is not known. Efforts to ascribe it to any known nearby nuclides which might have been produced by the irradiation of Tb^{159} have been unsuccessful; and it must, for the present at least, be regarded as the result of some unknown impurity in the terbium sample.

It is suggested that further experiments should be undertaken to resolve some of the apparently contradictory findings which have been reported to date in connection with the radioactive decay of Tb^{156}. An extensive series of irradiations at various energies below the maximum available of 64 Mev would establish experimentally the threshold for the production of Tb^{156} by the reaction $Tb^{159}(\gamma,3n)Tb^{156}$ and might help eliminate any concern over interfering radiation from other radioactive products. A search should be made for the 12.6-hour activity in several different irradiated samples of terbium metal and Tb_4O_7 to ascertain whether it is the result of an occasional impurity or a consistent product of the irradiation and whether transitions of other energies are found to exhibit the same half-life. Finally, gamma-gamma coincidence measurements should be helpful in separating activities with different origins but similar half-lives and in relating observed transitions to each other with sufficient certainty to allow the construction of a suitable level scheme.

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